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Highly birefringent, low-loss liquid crystals for terahertz applications

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With growing interest in switchable devices for the THz frequency range, there is a strong demand for liquid crystals (LC) exhibiting both a high birefringence and a low absorption. We present the refractive index and absorption coefficient of the liquid crystal mixtures 1852 and 1825 in the frequency range between 0.2 and 2.5 THz. Both mixtures are designed specifically for high birefringence Δn of 0.32 and 0.38, respectively, in the THz region. In addition, they show low absorption coefficients for both ordinary and extraordinary polarization. This low absorbance in combination with the high birefringence makes these LCs particularly well suited for applications in switchable devices for THz optics. © 2013 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [<http://dx.doi.org/10.1063/1.4808244>]

Terahertz technology is developing at a rapid pace.¹⁻³ While a decade ago bulky and expensive components mainly constrained terahertz systems to specifically equipped laboratories, new technical advances allow nowadays for applications in many different fields. Modern body scanners, for example, often operate in the lower THz frequency range. Other applications, for which it is anticipated that THz technologies will be increasingly implemented, span from plant physiology,⁴ investigations of the historical and cultural artifacts⁵ to monitoring industrial production processes.⁶ A field which will most certainly have an impact on our daily life is short-range wireless communication with THz waves, i.e., the wireless local area network (LAN) of tomorrow.⁷ Yet, a mature THz technology not only requires reliable and affordable sources and detectors but also devices which can be used to guide or manipulate THz waves. This includes modulators, phase-shifters as well as switchable reflectors and filters, all of which have been already demonstrated at least at a proof-of-principle level.⁸⁻¹⁰ Other devices which have not been demonstrated yet are directional couplers and phased arrays. Many of the already developed and future devices use or will use the birefringence provided by liquid crystals (LC).

The THz properties of many of-the-shelf liquid crystals have been studied in the past few years.¹¹⁻¹³ Most of these standard LCs, however, only show a relatively low birefringence in the THz frequency range. Recently, Wilk and co-workers systematically studied cyanobiphenyls (CB)^{11,14,15} and their mixtures.¹⁶ From these studies, it can be concluded that a pronounced π -electron system leads to a higher THz birefringence; in analogy to what is known from the optical frequency range. But in contrast to the visible frequency range where LCs are relatively transparent, they are moderately to strongly absorbent in the lower THz frequency range. The absorption predominantly arises from intramolecular vibrations involving the entire molecule. Yet, for efficient and in particular for thin THz devices, LC materials with a significantly higher birefringence, lower absorption, and

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low dichroism are crucial. Thinner components not only enable to switch the device faster, but by reducing the amount of LC material the cost-efficiency can be increased considerably. The challenge is, therefore, to design a LC system that not only exhibits a high birefringence but which also has favorable absorption properties.

Here, we present the refractive indices, birefringence, and absorption coefficients of the LC mixtures 1852 and 1825 in the THz frequency range. In contrast to the more random selection of LC materials investigated in the THz region so far, these structures were formulated and selected particularly for their low absorption in the THz frequency range, combined with a high birefringence and a low viscosity, what together insures a high Figure of Merit (FoM) value

$$\text{FoM} = \frac{\Delta n^2 \cdot k_{ii}}{\gamma}, \quad (1)$$

where k_{ii} is elastic constant: band, twist, or splay, and γ is rotational viscosity. Whereas the values of the absorption coefficients are typically very small at optical wavelengths and thus negligible for the description of the FoM, these values are more significant at THz frequencies and should not be neglected. We therefore prefer to include the loss properties according to

$$\text{FoM} = \frac{\Delta n^2 \cdot k_{ii}}{\gamma \cdot (\alpha_o + \alpha_e + |\Delta\alpha|)}. \quad (2)$$

This approach aims to determine a tailored LC system for fast applications at THz frequencies. The purpose of this comprehensive study is to further customize not only the optical parameters but also others such as the physical properties of the LCs; depending on the specific requirements of the respective application. The two mixtures are characterized using a standard THz time-domain spectrometer, allowing to extract both index and absorption simultaneously.

In order to achieve a high birefringence, two LC structures have proven to be very promising: Linear aromatic molecules, consisting of multiple benzene rings joined directly only via single bonds and LCs where the constituting phenyl rings are also joined via at least one triple bond. The triple bond is believed to be one of the main factors for the increased birefringence, due to the better conditions for conjugation between π electrons in the vicinity of benzene rings and an increase of the nematic features of the molecule. Both molecular structures characterize an easily polarizable π electron conjugated system along the long molecular axis, leading to a particularly high birefringence over a broad spectral range. However, such rigid core molecules have typically high melting points and strong smectic features, thus their potential for applications is limited. With a functionalization by a polar group (F, CN, NCS) in the terminal position and an alkyl group in the second terminal position; as well as by placing fluorine atoms in the lateral positions; the presence of the smectic phases can be strongly suppressed and the mixtures become nematic liquid crystals over a wide temperature range. This effect can be further increased if a cyclohexane ring is selected as one of the outer core rings, although the presence of fluorine atoms and cyclohexane rings can affect again the birefringence. It is thus crucial to achieve a good trade-off. Over the past few years, different isothiocyanato compounds were investigated, for example, by Catanescu and Chien¹⁷ as well as by our group.¹⁸⁻²⁴ These studies led to the selection of the following, particularly promising compounds, see Figure 1.

Note that X stands either for a fluorine or a hydrogen atom, and R stands for different terminations of the compounds which can be selected individually.²⁵⁻²⁷ In both mixtures, respectively, different variations for X and R for each of the 7 compounds can be present, depending on the particular composition. These compounds are very well suited to formulate a high birefringence mixture with improved performance for the THz range of the electromagnetic spectrum. Due to the absence of a polar bridge group in the molecular core, the intermolecular dipole-dipole coupling is small. Furthermore, they also do not dimerize as cyano-compounds, which leads to lower viscosity. It is believed that this contributes to the considerably low absorption at THz frequencies. Another contribution is ascribed to the presence of the triple bond in some of the compounds, which limits twisting oscillations of the benzene rings and out-of plane C-H librations. In addition, reorientation processes associated with intermolecular bonding may imply strong tensional oscillations that significantly affect the dielectric properties in the THz region.

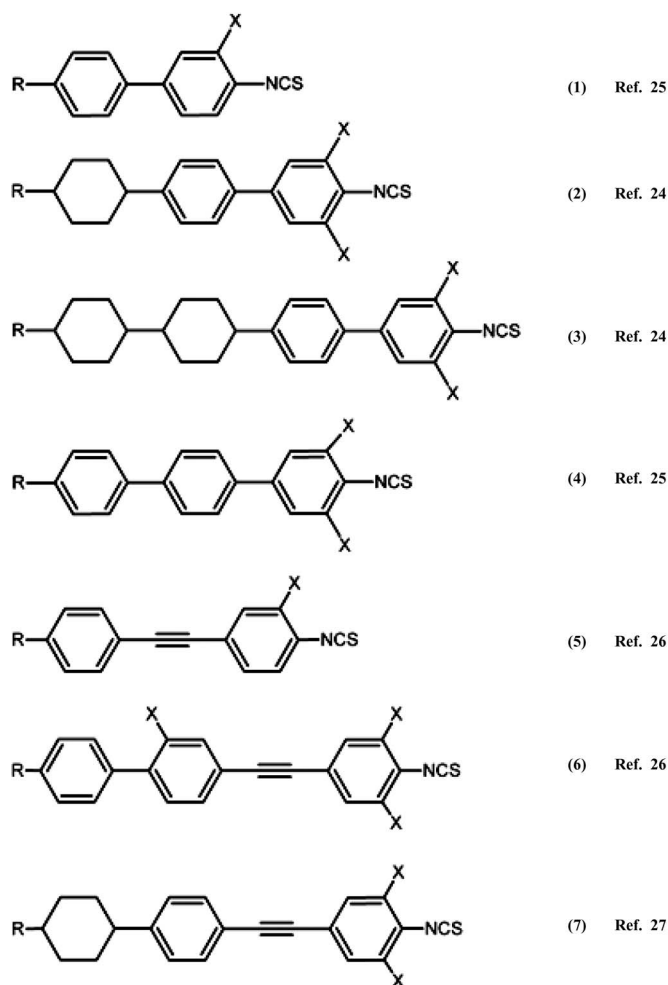


FIG. 1. Liquid crystal compounds used for the formulation of the investigated mixtures 1852 and 1825.

Two different mixtures with expected high birefringence were formulated 1825 and 1852. The mixture 1852 consists mainly of the compounds 1–4 (85 wt.%) and 5 (15 wt.%), while the mixture 1825 consists of the compounds, 5–7 (75 wt.%) as well as compounds 1 and 4 (25 wt.%), respectively. In order to limit the number of different conformational states and to lower the viscosity, molecules with short alkyl chains were selected. A very important characteristic of these mixtures is the fact that they both have an extraordinarily extended nematic range. The first mixture, i.e., 1825, is a nematic between -12°C and 136°C . The nematic range of the latter mixture, i.e., 1852, spans from -10°C to 152.6°C . This extended nematic range enables to apply these mixtures over a wide ambient temperature range; as in particular the birefringence is expected to change also only marginally within this range. The high clearing point of both mixtures is a further indicator for a very high birefringence.

The elastic and dielectric properties of the samples emphasize even more the potential of these structures for applications in fast switchable devices.²⁸ For the mixture 1852, at the temperature of 25°C , these parameters are: $\Delta\varepsilon = 15.3$, $\varepsilon_{\perp} = 4.1$ at 1.5 kHz, $K_{11} = 11.2$ pN, $K_{22} = 7.7$ pN, and $K_{33} = 31.01$ pN. For the mixture 1825, they are determined as $\Delta\varepsilon = 17.0$, $\varepsilon_{\perp} = 4.7$ at 1.5 kHz, $K_{11} = 12.5$ pN, $K_{22} = 7.4$ pN, and $K_{33} = 32.1$ pN.

The individual components of each of the mixtures were weighted and mixed, respectively. The mixtures were heated to their clearing point and after subsequent cooling to room temperature, they were filtrated through a Millipore system with durapore membrane filters with $0.22\ \mu\text{m}$ pore size.

A standard THz time-domain system as described in detail in Ref. 29 is used to characterize the LCs. A fs laser (Ti:Sa, Tsunami, Spectra Physics, 70 fs, 80 MHz, 780 nm) pulse drives and gates photoconductive antennas for generation and detection of the linear polarized THz radiation, respectively. By varying the time delay between generation and detection path of the system, the full temporal shape of the electric field can be recorded. The complete spectroscopic information is accessed via Fourier transformation.

A custom designed sample cell consisting of two 700 μm thick fused silica windows is used to hold the LCs. In order to assure a constant sample thickness of 1.3 mm, metal wires are employed as spacers between the windows. The distance between the wires, i.e., the width of the cell, is set to 15 mm. In addition, the metal wires act simultaneously as electrodes, thus allowing for the alignment of the LC molecules with a peak voltage of 1.5 kV at a modulation frequency of 1 kHz. The cell is kept at a constant temperature with a deviation of less than $\pm 0.1^\circ\text{C}$ for the entire measurements. However, different temperatures were set for the measurements of the different structures, owing to the specific lab settings at the different measurement campaigns. Due to the extraordinarily extended nematic phase of these LCs, the temperature dependence in this small temperature range is expected to be negligible. Mixture 1852 was measured at 23.6°C , while the temperature for 1825 was set to 20.5°C .

The entire THz beam path is kept under dry atmosphere in order to avoid artifacts arising from rotational lines of water molecules. THz pulses, propagating through nitrogen, i.e., with the cell removed, are recorded as reference $E_{\text{ref}}(t)$. The sample pulses $E_{\text{sam}}(t)$ are measured with the cell placed in the THz beam path. The LCs can be aligned perpendicular or parallel to the polarization of the THz radiation (ordinary or extraordinary polarization), respectively, by rotating the cell by 90° . After Fourier transform, the measured transfer function is given by

$$H_{\text{mes}}(\omega) = \frac{E_{\text{sam}}(\omega)}{E_{\text{ref}}(\omega)}. \quad (3)$$

For samples consisting only of one layer this is compared to the theoretical transfer function

$$H_{\text{theo}}(\omega) = (1 - r^2) \exp\left(-i(n - 1)d \frac{\omega}{c_0}\right) \sum_{m=0}^M \left(r^2 \exp\left(-i2nd \frac{\omega}{c_0}\right)\right)^m, \quad (4)$$

where r describes the losses due to reflections on the surfaces, n stands for the complex refractive index, d is the thickness of the sample, M is the number of multiple reflections within the sample, and c_0 is the speed of light in vacuum. By minimizing the difference of the two functions the optical parameters can be extracted. However, the sample cell consists of more than one layer that is why a more complex and highly precise parameter extraction algorithm is used,^{30,31} which also takes losses due to the windows and multiple reflections within the sample and the windows into account. This enables to minimize the uncertainties arising from thickness variations or misalignment of the cell.

The frequency dependent indices of refraction n_o and n_e and the corresponding birefringence Δn are shown for 1852 in Figure 2(a). Both indices decrease slightly over the accessible frequency range with a nearly featureless slope: n_e from 1.89 to 1.85, whereas n_o decreases from 1.57 to 1.55 from 0.5 THz to 2.5 THz. The resulting birefringence Δn is thus also homogeneous and nearly constant, varying by only 0.02 around an average of approximately 0.32. The uncertainty of the measurements for the refractive indices could be minimized to values below 0.003. The values of the extraordinary and ordinary index of refraction as well as the birefringence in the THz region are very similar to the values measured in the optical and infrared frequency ranges, indicating a relatively constant value over a broad spectral range,²⁸ as can be seen in Table I.

Figure 2(b) shows the absorption coefficient for both ordinary and extraordinary configurations, α_o and α_e , respectively. While for frequencies below approximately 0.9 THz, the extinction does not differ much whether the LCs are aligned parallel or orthogonal to the polarization of the THz radiation (small dichroism), for higher frequencies, α_e exceeds the values of α_o over the entire remaining accessible frequency range. This behavior is very different from what has been observed for the CBs and their mixtures¹⁴ for which a strong dichroism has been observed over the entire frequency range. Note that the extraordinary absorption coefficient exceeds the ordinary one for

TABLE I. Extraordinary and ordinary index of refraction, birefringence, and extraordinary and ordinary absorption coefficients of the mixture 1852 at 0.5, 1.0, and 2.5 THz. For comparison, the values obtained for the refractive indices and birefringence at 589 nm and 1064 nm are given.²⁸

	n_e	n_o	Δn	α_e (cm ⁻¹)	α_o (cm ⁻¹)
589 nm	1.86 ^a	1.53	0.33
1064 nm	1.81 ^a	1.52	0.29
0.5 THz	1.887	1.556	0.331	3	2.5
1.0 THz	1.886	1.559	0.327	8.5	7
2.5 THz	1.852	1.546	0.306	19	16

^a n_o and Δn were measured directly,²⁸ while n_e was calculated as $n_e = n_o + \Delta n$.

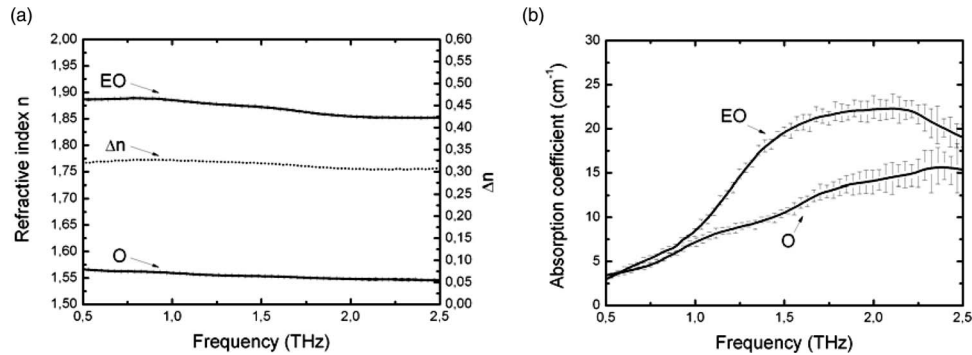


FIG. 2. (a) Index of refraction n for ordinary and extraordinary polarization and resulting birefringence Δn for the mixture 1852. (b) Absorption coefficient α of 1852 for both polarizations, respectively.

higher frequencies. The reason for this different behavior is not yet fully understood. We also observe a strong influence on the absolute values of the extraordinary absorption coefficient dependent on the choice of the terminal end group. Therefore, we hypothesize that due to the molecular structure, in particular the pronounced linearity of these LC molecules, dominant molecular vibrations in the extraordinary direction might be favored. Theoretical predictions of the far-infrared vibrational spectrum as well as ultrabroadband experimental spectroscopic data, which, for example, were recently reported for 5 CB,³² may help to verify this assumption. While other LC systems exhibiting a comparably high birefringence have been reported recently,³³ these structures typically suffer from high dichroism, thus limiting the application for switchable components significantly. The average absorption coefficient of 1852, i.e., the mean value of ordinary and extraordinary absorption coefficients, is therefore much lower than that of LCs with comparably high birefringence reported at THz frequencies so far.

The second mixture investigated in this study is 1825. The composition is selected purposely in order to achieve a particular high ratio of constituents with extended polarizable π electron conjugated system along the long molecular axis. The focus on such very long, multi-ring structures let us expect to achieve an even higher birefringence compared to the previous mixture while maintaining the low absorption. The values for the frequency dependent ordinary and extraordinary absorption coefficients are shown in Figure 3(b). Shape and absolute values of these parameters are very similar to those of mixture 1852. In particular, a very low dichroism is once more observed, even more accentuated for low frequencies.

The refractive indices n_o and n_e , and thus the birefringence Δn , shown in Figure 3(a), are yet noticeably higher. Considering in particular the low uncertainty achieved in this measurement, this birefringence Δn of the mixture 1852 is with approximately 0.38 the highest birefringence reported for the THz frequency range. It is worth noticing, however, that it is indeed the combination of these favorable parameters – high birefringence together with low absorption and low dichroism, which renders these mixtures as above all extremely suitable for applications in thin, switchable, and cost-efficient THz components. For comparison, the values obtained for the refractive indices

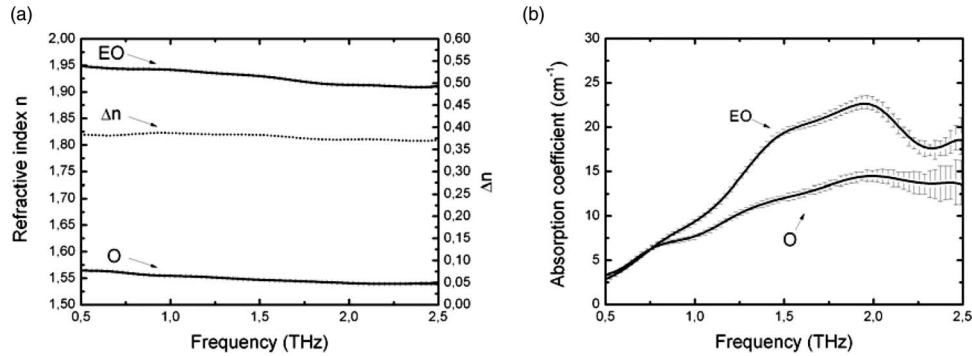


FIG. 3. (a) Index of refraction n for ordinary and extraordinary polarization and resulting birefringence Δn for the mixture 1825. (b) Absorption coefficient α of 1825 for both polarizations, respectively.

TABLE II. Extraordinary and ordinary index of refraction, birefringence, and extraordinary and ordinary absorption coefficients of the mixture 1825 at 0.5, 1.0, and 2.5 THz. For comparison, the values obtained for the refractive indices and birefringence at 589 nm and 1064 nm are included.²⁸

	n_e	n_o	Δn	α_e (cm ⁻¹)	α_o (cm ⁻¹)
589 nm	1.96 ^a	1.54	0.42
1064 nm	1.90 ^a	1.53	0.37
0.5 THz	1.948	1.565	0.383	2.8	3.3
1.0 THz	1.941	1.554	0.387	9.4	7.7
2.5 THz	1.911	1.540	0.371	19.0	13.7

^a n_o and Δn were measured directly,²⁸ while n_e was calculated as $n_e = n_o + \Delta n$.

and birefringence at 589 nm and 1064 nm are listed in Ref. 28, together with the values for n_e , n_o , Δn , α_e , and α_o at three selected THz frequencies.

These results also emphasize that the quest for new LC materials with various promising properties based on a comprehensive approach of customizing their design is successful. While the main focus lies definitely on enhancing the birefringence and decreasing the absorbance, other parameters determining the physical properties of the sample should not be completely neglected. The customized design of the LCs allows for controlling these physical parameters without impacting significantly on the excellent optical properties. The very high temperature range for which the LC material is in the nematic phase facilitates applications where a precise temperature control is not easily implementable.

We have shown that the mixtures 1852 and 1825, consisting of specifically selected compounds with a rigid molecular core and a long polarizable π electron conjugated system show a notably high birefringence at THz frequencies (Tables I and II). The careful selection of the constituents allows us to achieve values for the birefringence Δn in the low THz frequency region of up to 0.38. In addition, the LC mixtures exhibit only low absorption losses for both ordinary and extraordinary polarizations. Due to the high birefringence combined with the low absorption and low dichroism, these LCs are particularly well suited for thin and cost-efficient THz devices such as phase shifters, retarders, or beam shapers. The extended nematic phase of these LCs facilitates applications over a wide temperature range. Furthermore, we are able to customize the device in order to adjust the optical and physical parameters to the respective requirements.

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