Universal phase relation between longitudinal and transverse fields observed in focused terahertz beams

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\textbf{Abstract.} We directly observe longitudinal electromagnetic fields in focused freely propagating terahertz (THz) beams of radial and linear polarization. Employing electro-optic detection, which is phase sensitive, allows one to selectively detect longitudinal and transverse field components. A phase shift of $\pi/2$ between the transverse and longitudinal field components is revealed. This phase shift is of universal nature, as it does not depend on the mode, frequency and focusing conditions. We show that the universal phase relation is a direct consequence of the divergence-free nature of electromagnetic waves in vacuum. In the experiments, we observe the phase shift of $\pi/2$ for all frequency components of single-cycle THz radiation pulses of both radial and linear polarization. Additionally, we show that the longitudinal field of a radially polarized THz beam has a smaller spot size as compared with the transverse field of a linearly polarized beam that is focused under the same conditions. For field-sensitive measurements this property can be exploited even for moderate focusing conditions. Furthermore, the phase-sensitive detection of longitudinal electromagnetic fields opens up new possibilities to study their interaction with electronic excitations in semiconductor nanostructures.

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1. Introduction

In many textbooks, light is treated as a plane wave, that has a purely transverse character. In reality, except for azimuthally polarized beams, any beam of finite diameter exhibits longitudinal components [1]. In the focus the strength of the longitudinal fields relative to the transverse fields is enhanced. Longitudinal components were first observed in focused linearly polarized microwave beams [2]. While in that experiment the intensity of the longitudinal components was about 400 times smaller than the intensity of the transverse components, focusing radially polarized beams of visible light with high numerical aperture yielded intensities of the longitudinal components exceeding the intensity of the transverse components [3, 4]. Such beams exhibit a smaller spot size as compared to Gaussian beams of linear polarization [3, 5]. The observation of such strong and tightly focused longitudinal electromagnetic waves stimulated experimental [6–15] and theoretical [16–19] work on the generation of radially polarized beams and on their specific properties. Already in 1959, Richards and Wolf showed that the longitudinal field components of linearly polarized waves obey a phase relation of $\pi/2$ with respect to the transverse field components [20]; later Youngworth and Brown found the same result for radially polarized beams [21]. Experimentally, this relation has not yet been verified, since so far in all experimental studies the intensity has been measured. Hence those experiments cannot provide phase information. In the terahertz (THz) spectral range, field-resolved measurements revealing the phase information are a widely applied technique [22]. Furthermore, the method of electro-optic sensing employed to register the THz fields is capable of distinguishing between longitudinal and transverse field components. This has been demonstrated by detecting the longitudinal fields in the near field of a metallic tip [23].

In contrast with such THz fields associated with surface-plasmon polaritons, we investigate longitudinal THz fields of freely propagating waves of radial and linear polarization, and in particular, we measure the phase relation between longitudinal and transverse field components. We observe the predicted value $\pi/2$ for both radially and linearly polarized pulsed THz radiation. A symmetry argument is provided to reveal the universal character of this phase relation. The experimental results are compared with theory describing the beam properties beyond the paraxial approximation. A smaller spot size of the longitudinal components of a radially polarized beam compared with a standard Gaussian beam is observed even for moderate focusing conditions.

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Due to their symmetry, radially polarized beams offer a particularly intuitive access to understanding longitudinal fields and their phase relation with respect to transverse fields. In figure 1(a), a radially polarized continuous wave that is traveling along the z-direction and focused by a lens is depicted. The unfocused radially polarized wave has a donut-like intensity distribution and belongs to the class of Bessel–Gauss beams, which are solutions of the vector Helmholtz equation [24]. Already a simple geometric optics argument suggests that longitudinal fields should appear in the focus. As the wave fronts get tilted by the focusing element, the polarization vector exhibits components along the z-direction that add up in the focus (see figure 1(a)). The four small panels depict calculated field patterns in the focal plane for different phases of the wave. For \( \omega t = 0 \) the field distribution is purely transverse with the field pointing radially outwards\(^4\). A two-dimensional field pattern of this structure would require a charge in the center as a source of the field; hence, it is forbidden by Maxwell’s law \( \text{div} \ E = 0 \) in vacuum. In vacuum, the field pattern has to extend in the z-direction and a longitudinal field on the axis is required to provide an influx into the center of the radial pattern of the transverse components. Indeed, for the phase \( \omega t = \pi/2 \), the field is purely longitudinal over the whole focal plane with a pronounced maximum on the axis of propagation. The field is pointing in the negative z-direction and thus provides the required flux. For the phase \( \omega t = \pi (\omega t = 3/2\pi) \), purely transverse (purely longitudinal) patterns occur with inverted signs of the fields. Due to the symmetry of the transverse beam for \( \omega t = 0 \) and \( \omega t = \pi \) the maximum of the longitudinal field has to occur exactly for \( \omega t = \pi/2 \). We note that for phases that are not integer multiples of \( \pi/2 \), longitudinal and transverse fields occur simultaneously in the focal plane. The calculations for obtaining the beam patterns are based on the Fresnel diffraction integral formalism [25], which allows one to describe the fields in the vicinity of the focus also for cases where the paraxial approximation does not apply. Here the temporal dependence of the fields is separated and takes the simple form \( \exp(i \omega t) \). The solution of the spatial problem can be obtained in the form of integral equations for particular radiation modes. For radially polarized vector beams, the solutions for the transverse and longitudinal field components take the form

\[
E_{\text{trans}} = A \int_0^\alpha E_0(\theta) \cos^{1/2} \theta \sin 2\theta J_1(k\rho \sin \theta) \exp(ikz \cos \theta) \, d\theta, \tag{1}
\]

\[
E_{\text{long}} = iA \int_0^\alpha E_0(\theta) \cos^{1/2} \theta \sin^2 \theta J_0(k\rho \sin \theta) \exp(ikz \cos \theta) \, d\theta. \tag{2}
\]

Here \( k \) denotes the absolute value of the wave vector and \( \rho = (x^2 + y^2)^{1/2} \) the radial coordinate. \( \theta \) is the angle under which the electric field \( E_0 \) at the location of the focusing element is ‘seen’ at the location of the focus \( (z = 0) \) [21]. \( J_0 \) and \( J_1 \) are Bessel functions of the first kind and zeroth and first order, respectively. The constant \( A = (1/2)k f \) is proportional to the focal length \( f \). The patterns shown in figure 1(a) are calculated for focusing with a numerical aperture NA = 0.2. The electric field profile \( E_0 \) in front of the focusing element is described by a product of a

\(^4\) Of course the phase or, equivalently, \( t = 0 \), is only defined in a relative way. Here we set \( \omega t = 0 \) corresponding to the maximum of the transverse field pointing outwards and keep this convention throughout the paper. Equivalently, the phase \( \omega t = 0 \) corresponds to the maximum of the transverse field pointing in the positive x-direction for the linearly polarized beam.
Figure 1. Schematic presentation of focused beams of (a) radial and (b) linear polarization. In the small panels the calculated field distribution in the focal plane is depicted for different phases $\omega t$. Note that the direction of the transverse components is encoded in the false color in a different way for the two modes, namely in the radial direction for the radially polarized beam and along the $x$-axis for the linearly polarized beam. A scale bar indicates the wavelength $\lambda$.

Gaussian function and a Bessel function of first order, which is the field distribution of the lowest order Bessel–Gauss mode [21, 24]. The phase of the longitudinal and transverse field components in the focal plane ($z = 0$) is only determined by the real and imaginary factors in

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front of the integrals in equations (1) and (2), respectively. Hence the phase shift of \( \pi/2 \) between these components is independent of the wave vector or the focusing conditions.

Figure 1(b) shows a linearly polarized continuous wave focused under the same conditions as for the radially polarized wave. Again purely transverse \(^5\) (longitudinal) fields occur in the focal plane for \( \omega t = 0, \pi, 2\pi, \ldots \) (\( \omega t = \pi/2, 3/2 \pi, \ldots \)). The appearance of longitudinal fields even for linearly polarized beams can be understood as a consequence of \( \text{div} \, E = 0 \) for beams of finite diameter. For example, the transverse field pointing in the positive \( x \)-direction for \( \omega t = 0 \) requires on the right side of the pattern an outflux, which is provided by the longitudinal fields pointing in the positive \( z \)-direction for \( \omega t = \pi/2 \) (equivalent to \( \omega t = 3/2 \pi \)). Again the symmetry between the transverse pattern for \( \omega t = 0 \) and \( \omega t = \pi \) requires the maximum of the longitudinal fields to occur exactly at \( \omega t = \pi/2 \). We point out that this argument for the phase shift of \( \pi/2 \) applies for any mode, which exhibits both longitudinal and transverse components. The relative strength of the longitudinal fields compared to the transverse fields is much smaller for the linearly polarized beam as compared to the radially polarized beam (see different false color scales for \( E_{\text{long}} \) and \( E_{\text{trans}} \) in figures 1(a) and (b)).

For linearly polarized beams, integral equations can be derived for the electric field distribution in the vicinity of the focus. The field pointing in the \( x \)-direction and the longitudinal field are described by

\[
E_x = -iA \int_0^\alpha E_0(\theta) \cos^{1/2} \theta \sin \theta \left[(1 + \cos \theta) J_0(kr_p \sin \theta \sin \theta_P) + (1 - \cos \theta) J_2(kr_p \sin \theta \sin \theta_P) \cos 2\Phi_P\right] \exp(ikr_p \cos \theta \cos \theta_P) d\theta, \tag{3}
\]

\[
E_z = -2A \int_0^\alpha E_0(\theta) \cos^{1/2} \theta \, \sin^2 \theta \, \cos \Phi_P \, J_1(kr_p \sin \theta \sin \theta_P) \exp(ikr_p \cos \theta \cos \theta_P) d\theta. \tag{4}
\]

Here \( J_2 \) is the Bessel function of second order. Due to the lack of cylindrical symmetry the expressions are more complicated than in the case of radially polarized beams. Equations (3) and (4) are expressed in spherical coordinates \( r_p, \theta_P \) and \( \Phi_P \) instead of cylindrical coordinates \( \rho \) and \( z \) applied to equations (1) and (2). Similarly to equations (1) and (2) only the prefactors define the phase of the fields in the focal plane \( (\theta_P = \pi/2) \). For calculating the beam profiles depicted in figure 1(b) a Gaussian profile in front of the focusing optics and a numerical aperture \( \text{NA} = 0.2 \) are employed.

3. Experimental method

In our experiment, we generate radially and linearly polarized single-cycle THz pulses by exciting photoconductive structures with near-infrared femtosecond laser pulses \( (\text{wavelength } 800 \text{ nm, repetition rate } 78 \text{ MHz, duration } 50 \text{ fs and pulse energy } 8 \text{ nJ}) \). A microstructured interdigitated electrode structure, consisting of linear stripes in the case of the emitter for linearly polarized radiation \([26]\) and concentric rings for the radial polarization \([12]\),

\(^5\) We note that there are also transverse components along the \( y \)-direction in the focal plane (not shown). For the focusing conditions chosen here they are about four orders of magnitudes weaker as compared the components in the \( x \)-direction. Since both transverse components are in phase, the components in the \( y \)-direction also exhibit the \( \pi/2 \) phase shift with respect to the longitudinal components.
respectively, is patterned on semi-insulating GaAs. The divergent THz beam is collimated and refocused by a pair of polymethylpentene (TPX) lenses with a focal length of 75 mm. Two 200 μm thick ZnTe crystals are placed in the focal plane alternately for electro-optic sampling (EOS). Changing the time delay between the pulse exciting the THz emitter and the probe pulse allows for registering THz transients. By scanning the detection unit, consisting of the ZnTe crystals and the optical components to read out the near-infrared polarization state, beam profiles along the x-direction (for y = 0) are measured in steps of 0.1 mm.

Depending on the orientation of the ZnTe crystal, the EOS technique is sensitive to either transverse or longitudinal THz fields. A (110)-oriented crystal is employed for registering the transverse field components, while a (100)-oriented crystal is used for detecting the longitudinal field components [27]. The orientation of the crystal axis is confirmed by x-ray diffraction. The high selectiveness of the detection with regard to longitudinal or transverse fields is highlighted by measurements of THz-signal dependence on the azimuthal orientation of the crystals (cf figure 2). The dependences clearly reflect the different symmetries of the crystal planes. The measured data agree with the predicted dependence, indicating that the method is well suited for selectively detecting the longitudinal and transverse components. The small deviations are due to slightly imperfect orientations of the crystals.
4. Experimental results for radially polarized terahertz (THz) pulses

In figure 3, the measured transverse and longitudinal THz fields are shown for a radially polarized beam. The transverse fields clearly show the feature of a radially polarized single-cycle THz pulse: a main THz half-cycle with the field pointing radially outwards (i.e. positive THz fields for $x > 0$ and negative THz fields for $x < 0$) around zero time delay, preceded and succeeded by weaker half-cycles with the field pointing radially inwards. The longitudinal field components are strongest around $x = 0$, i.e. on the axis of propagation where the transverse field vanishes. In figures 3(b) and (d), THz transients at different spatial positions are shown. The transverse components for $x = 0.3$ mm and $x = -0.3$ mm are typical half-cycle pulses of opposite sign. The longitudinal field at $x = 0$ has the shape of a single-cycle pulse with a zero crossing at zero time delay. The change in pulse shape between the longitudinal and transverse fields is a natural consequence of the phase shift for a spectrally broad pulse. For zero time delay, the longitudinal field is zero for all values of $x$; in this sense it can be referred...
Figure 4. Phase difference of the longitudinal and transverse fields. The black squares are data measured for $x = 0.1$ mm and the green dots those measured for $x = 0.2$ mm. The dashed line indicates the value $\pi/2$. In the inset the amplitude spectra of the longitudinal (red) and transverse (blue) components measured at $x = 0.2$ mm are depicted.

to as a sine-shaped pulse (and the transverse field as cosine-shaped pulses). This indicates that all frequency components of the single-cycle THz pulse obey the phase shift of $\pi/2$ between the longitudinal and transverse fields in the entire focal plane. For demonstrating this more quantitatively, the frequency-dependent amplitude and phase are calculated by Fourier transformation of the THz transients. As one can see in the inset of figure 4, the spectral content of the longitudinal and transverse components is similar for a fixed location $x$. The phase difference $\Delta \Phi = \Phi_{\text{trans}} - \Phi_{\text{long}}$ between the longitudinal and transverse components is found to be $\pi/2$ for all reasonably strong frequency components.

5. Experimental results for linearly polarized THz pulses

We now investigate the much more common mode of linearly polarized THz beams. The transverse components are characterized by a half-cycle pulse with a maximum for $x = 0$ and $t = 0$ (figure 5(a)). While no longitudinal fields are observed on the axis of propagation, significant longitudinal fields occur at around $|x| = 0.3$ mm. The measured longitudinal fields exhibit a single-cycle form with negative (positive) fields for negative time delays and positive (negative) $x$-values. This is in accordance with the theoretical expectation (see figure 1(b) for $\omega t = 3/2\pi$). Similarly, the observed longitudinal fields for positive time delays correspond to the pattern presented in figure 1(b) for $\omega t = \pi/2$. Another scan was performed in the $y$-direction, i.e. perpendicular to the direction of polarization. In this case a similar transverse field profile but vanishing longitudinal fields were observed (not shown). Comparing figures 5(a) and (b) we find that the maximum of the transverse field occurs at the time when the longitudinal field is zero for all values of $x$. Hence, also all frequency components of the focused linearly...
polarized beam are characterized by a phase shift of $\pi/2$ between the transverse and longitudinal fields. We note that the X-like pattern seen in the diagrams are signatures of diffraction limited broadband pulses. Such patterns have been observed for single-cycle THz pulses emitted from antennas [28] and are also seen in calculated patterns for radially polarized radiation from accelerator-based sources [13].

6. Comparison of spot sizes of longitudinally and radially polarized beams

In this section, the results of the experiments involving radially and linearly polarized beams are compared with each other and with theory. In figure 6, beam profiles for the two field components are depicted. The profiles are selected for the times where the corresponding component reaches maximum amplitude. The calculations, based on equations (1)–(4),
Figure 6. Beam profiles for (a) radially and (b) linearly polarized beams. The red squares correspond to the measured longitudinal fields and the black circles to the transverse fields. The lines represent calculated beam profiles (see the main text).

are performed for $\lambda = 0.3$ mm. The full-width at half-maximum of the Gaussian function characterizing the beams in front of the focusing element is 30 mm for both types of beams. This resembles the situation for the central wavelength of the single-cycle pulses studied in the experiment [12]. This calculation reproduces the measured fields very well (see figure 6). Note, however, that in the experiment the radiation is broadband. In both the measured and calculated beam profiles, the full-width at half-maximum $w$ of the longitudinal field of the radially polarized beam is smaller than the width of the transverse component in the case of linearly polarized radiation. While this effect is expected, there is an important difference to many experiments in the visible spectral range, where only the total intensity is recorded. In the latter case, the tighter focusing of radially polarized beams is only observed for very large numerical apertures, since it requires the longitudinal component to be stronger than the transverse components. In our case of direction-sensitive detection of the fields, however, the narrower beam profile can already be observed for moderate focusing, where the transverse components still exceed the longitudinal components.
7. Discussion of implications and outlook

Finally, we discuss some implications of our findings. The knowledge of the phase relation between longitudinal and transverse fields can be applied to detect these components selectively even when the applied technique is insensitive to the field orientation. An example of such a technique is the Kerr–Shutter method [29]. For wavelengths in the visible or UV range, where temporal gating becomes difficult due to the short period of the waves, interferometric techniques may be applied in order to exploit the spatial separation of the longitudinal and transverse fields caused by the universal phase shift. This may enhance the spatial resolution, for example, in nanolithography. Moreover, our technique can be employed to study fundamental aspects of light–matter interaction. For the intensity-based experiments in the visible range it has been debated controversially whether a photodetector can register the longitudinal field in the center of a radially polarized mode, since the latter is accompanied by a vanishing magnetic field component and thereby a vanishing Poynting vector [4, 29]. The possibility to register the amplitude and phase of transverse and longitudinal THz field components should allow one to gain insights into this interesting issue, e.g. by probing the transmission through quantum wells. Intersubband transitions in quantum wells require a field component perpendicular to the quantum-well plane. Hence, electrons in quantum wells will interact strongly with the longitudinal field when the quantum wells are placed on the electro-optic sensor crystal. With accelerator-based sources [13] capable of generating radially polarized fields in the MV cm$^{-1}$ range, even the high-field regime can be investigated.

8. Summary

In conclusion, by directly recording the transverse and longitudinal field components of single-cycle THz pulses, we have provided insights into the spatial and temporal structure of the fields. The observed phase difference of $\pi/2$ between longitudinal and transverse fields, which can be concluded from a symmetry argument, is of universal nature, i.e. it does not depend on frequency, the type of mode or focusing conditions. It can be applied to selectively detect longitudinal or transverse field components even when the detection mechanism itself is not sensitive to the field direction. This may lead to a better understanding of light–matter interaction in the case of the vanishing Poynting vector.

Note added in proof. After the submission of this paper for publication, complementary data on longitudinal fields observed in radially polarized THz beams generated by a segmented nonlinear optical crystal have been published [30].

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