Running electric field gratings for detection of coherent radiation

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A technique for the room temperature electro-optic detection of coherent low-frequency radiation, typically in the far infrared or terahertz (THz) range, is proposed. It relies on a longitudinally running electric field grating probed by phase-matched Raman–Nath diffraction of a higher frequency wave. The expected probe wave diffraction intensity is background-free and is proportional to the desired signal intensity. As an example, we present model calculations for the probing of monochromatic THz radiation using the organic crystal 4-N,N-dimethylamino-4'-N'-methyl-stilbazolium 2,4,6-trimethylbenzenesulfonate as nonlinear optical sensor material, for which new measurements of the refractive index and absorption up to a frequency of 11 THz are presented.

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1. INTRODUCTION

Approaches based on nonlinear optics are among the most valid alternatives to conventional thermal sensors for the detection of far infrared or terahertz (THz) radiation, such as bolometers, Golay cells, or pyroelectric detectors [1], some of which require cryogenic temperatures. Nonlinear optics brings about a faster response, permits operation at room temperature, and offers the interesting feature that the signature of the long wavelength radiation can be transferred to a wave at much shorter wavelength, easily detected by means of standard photodiodes or photomultipliers. This is true both for nonlinear techniques based on frequency conversion [2–5] and for those based on electro-optics, as used for instance in electro-optic time-domain spectroscopy [6]. Importantly, in the latter case the electro-optic phase shift is proportional to the local electric field of the electromagnetic wave under study (signal wave). An interesting situation is the one where this wave is spatially inhomogeneous, e.g., tightly focused. In this case the spatial variation of the cumulated phase shift on a copropagating probe wave can produce a distortion of its wavefront on top of the average phase shift. This distortion leads in general to a slight focusing or defocusing of the probe wave. This useful phenomenon was exploited by Schneider and coworkers [7–9] to develop a simple and effective method to detect ultrashort (nearly single cycle) broadband THz pulses, also known as terahertz-induced lensing (TIL). However, extension of this method for the detection of radiation with significantly longer pulses and weaker peak intensity is difficult, an important drawback being the fact that the weak probe wave intensity variation is measured over a strong background that exists also in the absence of the signal wave.

Here we propose and analyze an alternative method that faces this drawback, while still exploiting a spatial variation of the signal wave electric field. The proposed technique is based on the creation of a running electric field grating and can work for a sufficiently narrow-bandwidth coherent signal wave able to create few interference fringes. The signal wave grating is probed by a wave being velocity-matched to the longitudinal speed of the grating and leading to diffraction in angular directions where a potentially background-free measurement is possible. In Section 2 we describe the concept and the underlying physics and analyze its requirements. In Section 3 we propose a compact implementation based on a Lloyd type interferometer with incidence either through the front surface or an oblique side surface of the nonlinear sample. Finally, in Section 4 we give an application example related to monochromatic THz wave detection based on the organic crystal 4-N,N-dimethylamino-4'-N'-methyl-stilbazolium 2,4,6-trimethylbenzenesulfonate (DSTMS). Thanks to new measurements of the THz refractive index

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of this crystal, we predict the necessary phase-matching configuration and we estimate the achievable performance in this context.

2. PHYSICAL CONCEPT

As shown in Fig. 1, the signal wave of angular frequency $\omega_s$ is split into two parts and enters a nonlinear optical material under the external angle $\theta_{s,ext}$, while the auxiliary probe wave at frequency $\omega_p = 2\pi c/\lambda_p$ enters the same material under an angle $\theta_{p,ext}$. Even though from physical principles there is no limitation on the values of the two involved frequencies, from a practical point of view $\omega_p$ should be such that this wave is detectable by means of conventional detectors, e.g., a visible or infrared photodiode.

Let us consider for simplicity that the two signal beams, modeled as plane waves, enter the sample symmetrically with respect to the longitudinal $z$-axis. The electric fields of the corresponding waves are $\vec{E}_1(\vec{r}, t) = E_{01}/2 \hat{\xi} \exp[i(\omega_p t - k_1 \cdot \vec{r})]$ and $\vec{E}_2(\vec{r}, t) = E_{02}/2 \hat{\eta} \exp[i(\omega_p t - k_2 \cdot \vec{r})]$. Here $\hat{\eta}_{1,2}$ are the unit vectors along the electric field direction of the two waves, $\hat{\xi}_{1,2} = (\cos \theta, 0, \pm \sin \theta)$ for $p$-polarized waves and $\hat{\eta}_{1,2} = (0, 1, 0)$ for $s$-polarized waves. The corresponding wave vectors are $k_{1,2} = k_0(\pm \sin \theta, 0, \cos \theta)$, where $\theta$ is the internal angle of propagation with respect to the surface normal (z-axis). The coherent superposition of the two waves $\vec{E}(\vec{r}, t) = \vec{E}_1(\vec{r}, t) + \vec{E}_2(\vec{r}, t)$ leads to

$$\vec{E}(\vec{r}, t) = E_0 e^{i(\omega_p t - k_1 \cdot \vec{r})} e^{-\alpha z \left(1/2 \cos \theta \right)} \vec{\tilde{x}}(z),$$

where we have taken into account the effect of a possible attenuation of the signal wave upon propagation by means of an absorption constant $\alpha$, for its intensity. Here $\vec{\tilde{x}}(z)$ is a complex vector that depends on the transversal $x$-coordinate and takes into account the local amplitude and direction of the combined electric field. It is given by $\vec{\tilde{x}}(z) = (\cos \theta \cos(k_0 x), 0, i \sin \theta \sin(k_0 x))$ for $p$-polarization and by $\vec{\tilde{x}}(z) = (0, \cos(k_0 x), 0)$ for $s$-polarization, its modulus is always $|\vec{\tilde{x}}(z)| \leq 1$. Equation (1) represents an electric field grating modulated along the $x$-coordinate. The phase of this grating is running along the longitudinal $z$-coordinate at the speed $v_{sp} = \omega_p/k_0 = \omega_s/(k_0 \cos \theta_s)$, where $c$ is the speed of light in vacuum and $n_s(\theta_s)$ is the refractive index seen by the signal wave.

If the material where the waves cross is optically nonlinear, the electric field grating can translate into a refractive index grating. Here we focus only on materials exhibiting optical nonlinearities of second order, for which the change in refractive index is linear in the local electric field. In principle also third order (Kerr-type) nonlinearities can be used, but this effect is generally much weaker and would therefore be useful only for detecting very intense signals. By an appropriate linear electro-optic tensor element the running electric field grating of Eq. (1) gives rise to a running refractive index grating $\Delta n(\vec{r}, t)$, which is able to influence the propagation of the probe wave through the sample. However, a significant cumulation of the local phase retardation on the probe wave can occur only if the longitudinal phase velocity of this wave equals or nearly equals the longitudinal velocity of the grating phase. This leads to following phase-matching condition:

$$n_s(\theta_s) \cos \theta_s = n_p(\theta_p) \cos \theta_p,$$

where $n_s$ and $\theta_s$ are the refractive index and the internal angle for the auxiliary probe wave, respectively. For later use, we define also a phase mismatch parameter as

$$\delta n \equiv n_p(\theta_p) \cos \theta_p - n_s(\theta_s) \cos \theta_s.$$

As seen in Eq. (2), the phase-matching condition can be advantageously adjusted to a certain extent by changing the angles $\theta_s$ and $\theta_p$.

We are interested here in the diffraction of the auxiliary probe wave of vacuum wavelength $\lambda_p$ in the Raman–Nath regime [10]. This diffraction is a result of the cumulated transverse modulation of its phase upon propagation along the running refractive index grating that possesses the transverse grating vector $k_x = k_0 \sin \theta_s = (2\pi/\lambda_p) n_p(\theta_p) \sin \theta_p$. Provided that the condition of Eq. (2) can be fulfilled, the ideal situation that gives the maximum phase modulation is the one where the probe wave propagates along the $z$-axis ($\theta_s = 0$, perpendicularly to the grating vector. Let $\phi(\xi, z) = \phi_0(L, t) \cos(k_x z)$ be the $x$-modulated phase shift experienced by such an incident plane probe wave upon propagation through the above running grating of thickness $L$. This quantity can be determined by inserting the running grating $\Delta n(\vec{r}, t)$ into the wave equation; its amplitude is a solution of the integral

$$\phi_0(L, t) = \frac{2\pi}{\lambda_p} \Delta n_0(t) \int_0^L \cos \left(\frac{2\pi}{\lambda_p} \delta n z \right) e^{-\alpha z} \, dz,$$

where $\Delta n_0(t)$ is the amplitude of refractive index grating at the time $t$ corresponding to the entrance of the probe wave into the sample (at $z = 0$). The projected amplitude absorption constant $\alpha_s$ in the above equation is defined as $\alpha_s \equiv \alpha_e/(2 \cos \theta_e)$. Equation (4) leads to

$$\phi_0(L, t) = \frac{2\pi}{\lambda_p} \Delta n_0(t) \left[ e^{-\alpha_s z} + \xi^2 \right]$$

$$\cdot \left[ e^{-\alpha_s L}(\xi \sin \xi L - \alpha_s \cos \xi L + \alpha_s) \right],$$

where the quantity $\xi$ is defined as $\xi \equiv 2\pi \delta n/\lambda_p$. If the absorption for the signal wave can be neglected, Eq. (5) reduces to the simpler expression

$$\phi_0(L, t) = \left( \frac{2\pi}{\lambda_p} \Delta n_0(t) L \right) \sin \left( \frac{2\pi}{\lambda_p} \delta n L \right),$$

Fig. 1. Configuration for the detection of the signal wave via an auxiliary probe wave diffracted by a longitudinally running grating in the nonlinear material. If the waves are pulsed, synchronization of the pulses is required.
where the cardinal sine factor disappears at perfect phase matching. The above amplitude is modulated at the frequency $\omega_0$ of the signal wave according to Eq. (1). In the Raman–Nath diffraction regime, the transversely phase-modulated probe wave leads to several diffraction orders in the transmitted far-field. The diffraction efficiency into the order $n$ is given by $\eta_n(t) = |f_n(\phi_n(t))|^2$, where $f_n$ is the Bessel function of the first kind of order $n$. For small values of the phase shift amplitude ($\phi_0 \ll 1$) one obtains for the diffraction efficiency into the first order (order +1 or −1)

$$\eta_{\pm 1}(t) \approx \left(\frac{\phi_0}{2}\right)^2 \approx \left(\frac{\pi}{\lambda_a} \Delta n_0(t) L \right)^2 \sin^2\left(\frac{2\pi}{\lambda} \delta n L\right).$$  (7)

The second equality in the above expression holds only for negligible absorption ($\delta a, L \ll 1$). Since $\Delta n_0$ is proportional to the electric field, the above diffraction efficiency is expected to scale linearly with the intensity of the signal wave. Also, due to the quadratic term in Eq. (7), the diffracted wave intensity is temporally modulated in amplitude at twice the frequency $\omega_0$ of the signal wave.

Let us now discuss the optimum conditions with respect to the thickness of the nonlinear sample. First we mention that if the absorption of the signal wave is very strong, the interaction distance will be controlled by its effective absorption length $L_a = 1/\delta a$. In this case, the use of thick samples is possible provided that $L_a$ does not contradict any of the arguments given below. By considering now the case of weak absorption, we see from Eq. (2) that the diffraction efficiency falls to half its maximum value if the argument of the $\sin^2$ function in Eq. (2) equals 1.39. Therefore, for a given mismatch $\delta n$ the grating thickness should not exceed about $L_{\text{max}} \approx 0.2215 \lambda / \delta n$, which is of the order of the signal wave wavelength, unless the mismatch parameter $\delta n$ is significantly smaller than 0.2. However, even in the case where the phase-matching condition Eq. (2) is well satisfied, the thickness $L$ should not be excessively large. This is to avoid that the diffraction process gets into the Bragg regime (thick grating). Indeed, satisfying simultaneously the Bragg condition and the relation Eq. (2) would be very difficult to achieve. The condition to be satisfied for the Raman–Nath diffraction regime was given in [11,12] and reads for our case as

$$\frac{\Delta n_0}{2n} k_0^2 L^2 = \frac{2\pi}{\lambda} \frac{\Delta n_0}{n} \frac{L^2}{\Lambda} \leq 1,$$  (8)

where $n$ is the average refractive index seen by the probe wave and $\Lambda = 2\pi / k_0$ is the grating period. It is unlikely that the ratio $\Delta n_0 / n$ will much exceed a value of the order of $10^{-5}$, so that the ratio $L/\Lambda$ can be up to an order of about 100 according to this criterion.

The above limitations on the thickness of the grating holds for perpendicular incidence of the probe wave ($\theta_a = 0$). In the case of oblique incidence, even under phase matching, the amplitude of phase modulation $\phi_n(L, t)$ is decreased due to the fact that the phase corruption accumulated by the probe wave is spatially averaged as a result of propagation in a region with different (and eventually opposite) refractive index contrasts. This issue was already recognized in the original work by Raman and Nath [10]. A reasonable condition to avoid this effect may be given by requiring that the side drift of the probe wave should not exceed about half a grating period over the propagation distance through the sample, that is $\tan \theta_a < \Lambda / 2L = \pi / (k_0 L)$ or, equivalently, $L < \Lambda / (2 \tan \theta_a)$. Under phase matching, this gives the strongest limitation on the usable grating thickness already for an internal angle $\theta_a$ of a few tenths of a degree. Note, however, that secondary (but weaker) maxima of the cumulated phase corrugation are found for specific angles $\theta_a$ exceeding the above limit and corresponding to the conditions $\tan \theta_a = (3 + 2p) \Lambda / 2L$, where $p$ is a positive integer. This relaxes somehow the last inequality if the use of larger probe wave angles is needed for fulfilling Eq. (2). The amplitude of the phase modulation $\phi_n(L, t)$ is reduced by roughly a factor of 5 for $p = 0$ with respect to the case of normal incidence ($\theta_a = 0$), and by nearly a factor of 10 for $p = 1$.

**3. SINGLE SIGNAL BEAM CONFIGURATION**

In certain wavelength ranges, the use of the beam splitter for the signal wave depicted in Fig. 1 may be impractical. A more compact alternative approach is the use of a Lloyd-type interferometer, as shown in Fig. 2. In this case only one signal wave is directed to the nonlinear optical detection sample and the second wave is created by a mirror in contact with the sample and/or by total reflection at its side surface. The probe wave propagates parallel (or nearly parallel) and very close to this side surface. If the signal wave enters through the front surface [Fig. 2(a)], the phase-matching condition Eq. (2) can be fulfilled for $\theta_a = 0$ provided that the refractive index $n_s$ of the signal wave is in the range

$$n_a \leq n_s \leq \sqrt{n_a^2 + 1},$$  (9)

as limited by the refraction at the entrance surface. This range holds also for the case of the two-wave interference of Fig. 1 and is depicted in Fig. 3 as the violet shadowed area. Due to the limitation of the range of $n_s$, in Eq. (2), the front surface configuration of Fig. 2(a) is suitable for materials for which the refractive indices at the frequencies $\omega_0$ and $\omega_s$ do not differ too much, which may be the case for several organic crystals even if $\omega_0$ is in the THz range. If the difference of the refractive indices $n_a$ and $n_s$ is larger, one may still fulfill Eq. (2) for $\theta_a = 0$ if the signal wave is incident through a wedged side surface, as shown in Fig. 2(b). In this case, the allowed values for the refractive index $n_s$ depend on the wedge angle $\beta$ and we have

$$\frac{\sqrt{n_a^2 - 2n_a \cos \beta + 1}}{\sin \beta} \leq n_s \leq \frac{\sqrt{n_a^2 + 2n_a \cos \beta + 1}}{\sin \beta}.$$  (10)
The corresponding range for \(n_s\) is depicted in Fig. 3 as the orange shaded area. This approach may allow one to employ nonlinear materials for which the dispersion of the refractive indices between the frequencies \(\omega_s\) and \(\omega_a\) is rather large, as expected for most inorganic crystals. Note that the inequality Eqs. (9) and (10) assume ordinary (angle-independent) refractive indices for the signal wave. An extension to the case of an extraordinary polarization is straightforward. Note also that Snell’s law prevents the possibility to fulfill Eq. (2) under side illumination of a rectangular nonwedge sample (\(\beta = 0\)).

The wedged configuration of Fig. 2(b) requires a minimum propagation distance of the signal wave in the sample before reaching the interaction region. If the wedge angle \(\beta\) is not too large, this distance, corresponding to the average sample width, may be kept below 1 mm. Nevertheless, in the case where the absorption constant \(\alpha_s\) is such that the signal wave is already strongly depleted over such a distance, the two-beam configuration of Fig. 1 or the front surface one-beam configuration of Fig. 2(a) have to be preferred.

4. APPLICATION EXAMPLE

Even though the theoretical treatment does not depend on the values of the frequencies \(\omega_s\) and \(\omega_a\), we illustrate the concept by using two distinct lasers as pumps, dual wavelength optical parametric oscillators (OPO) [19], or compact dual frequency solid-state or fiber laser sources [20–23]. We will now check...
whether the THz radiation can in principle be detected with the approach presented in this work by recycling part of one of the near infrared pump waves as the auxiliary probe wave at the detection stage, which gives a potentially compact ensemble.

Let us therefore take \( \nu = \omega / 2\pi = 4 \) THz for the signal frequency and \( \nu_a = \omega_a / 2\pi = 222 \) THz (\( \lambda_a = 1350 \) nm) for the probe frequency. Inspection of the situation for DSTMS leads to an optimum configuration if both signal and probe waves are polarized along the (vertically oriented) polar \( a \)-axis of the crystal, which permits us to activate the largest high-frequency electro-optic coefficient \( r_{111} \approx 37 \) pm/V of DSTMS that was evaluated for the wavelength of 1.9 \( \mu m \) [16]. The refractive indices needed to determine the optimum configuration are therefore \( n_1(\omega_a) = 2.091 \) [16] and \( n_1(\omega_s) \approx 2.24 \) (Fig. 4). Since \( n_1(\omega_s) > n_1(\omega_a) \) and the inequality Eq. (9) holds, the phase-matching condition of Eq. (2) can be fulfilled here for the ideal case where the IR probe wave is normal to the grating vector and all waves enter through the front surface. This is shown in Fig. 5(a), which shows the expected normalized diffraction efficiency as a function of the internal angle \( \theta_s \) of the incident THz waves, under the condition \( \theta_r = 0 \). It is seen that the choice of the phase-matching angle \( \theta_s \) is not too critical, since the curves show a rather broad angular aperture (about 5 deg full width at half-maximum for a sample with \( L = 500 \) \( \mu m \)). The thick red and blue curves in Fig. 5(a) are determined by explicitly considering the effect of the signal wave absorption (\( \alpha_s = 5 \) mm\(^{-1}\)) on the output phase corrugation amplitude in Eq. (5). For comparison, the case of no absorption is also shown (thin green curve) and gives the reference for the diffraction efficiency normalization. Figure 5(b) compares the angular rocking curve for \( L = 250 \) \( \mu m \) and \( \nu_s = 4 \) THz of case (a) with the corresponding curves for the same probe wavelength but other THz frequencies, i.e., \( \nu_s = 2 \) THz (\( n_1 \approx 2.22, \alpha_s = 4 \) mm\(^{-1}\)) and \( \nu_s = 6 \) THz (\( n_1 \approx 2.22, \alpha_s = 15 \) mm\(^{-1}\)). This shows that a nearly identical geometrical configuration of the detector (moderate change of \( \theta_s \)) can be employed to detect the coherent radiation in a rather broad spectral range, provided that the dispersion of the relevant refractive indices in this range is not too strong, which is the case for the example of DSTMS.

It is useful to get some order of magnitudes for the expected effects. For the example depicted in Fig. 5(a), the optimum grating period is \( \Lambda = 93 \) \( \mu m \) and the external diffraction angle of the ±1 order with respect to the transmitted zeroth order is \( \lambda_s / \Lambda \approx 14 \) mrad. If we assume that the signal wave has 1 W peak power (e.g., 10 ns long pulses with roughly 10 nJ energy) concentrated over a surface of 1 mm\(^2\) we obtain an electric field of the order of 180 V/cm and a maximum refractive index modulation \( \Delta n_0 \approx 3 \cdot 10^{-6} \) in the case of DSTMS. For \( L = 500 \) \( \mu m \) and considering the THz absorption this leads to \( \phi_s \approx 0.004 \) in Eq. (5) and a diffraction efficiency of \( \eta_{\pm 1} \approx 0.0004\% \) into the first Raman–Nath order. Even though this value may seem small, the diffraction spots appear over an otherwise dark background in the absence of scattering and should therefore be easy to measure by appropriate conventional detectors responding at the probe wave frequency. It appears reasonable that a diffraction efficiency at least two orders of magnitude lower would be still detectable, corresponding to electric fields of a few tens of V/cm or a peak intensity of a few mW/mm\(^2\). Note also that for the above parameters we have \( 2\pi^2(\Delta n_0/n)L^2 / \Lambda^2 \approx 0.001 \leq 1 \) and we are therefore well inside the Raman–Nath diffraction regime. In general, the major source of noise for the detection is likely to be given by linearly scattered light at the probe wavelength \( \lambda_s \), which magnitude depends on the crystal and surface quality, as well as on the wavelength \( \lambda_s \) and the expected diffraction angle. If necessary, an additional slow external modulation of the signal wave might be used to improve the discrimination against this scattering using lock-in techniques.

The above application example can be extended to various other nonlinear materials as detectors. For instance, one might use instead of DSTMS the well-known and closely related DAST crystals, 4-N,N-dymethylamino-4’-N-methyl-stilbazolium tosylate [24–26]. Let us consider the THz frequency \( \nu_s = 2 \) THz and the probe wavelength \( \lambda_s = 1.5 \) \( \mu m \), which in DAST represent a nearly phase-matched pair at the generation stage [25]. With the IR and THz refractive

![Diagram](image-url)
indices given in [24] and [26] one obtains an optimum configuration of the type of Figs 1 and 2(a) with an internal angle $\theta_i = 19.7$ deg. For the same intensity as in the example above one would expect here a diffraction efficiency $\eta_{x,1} \approx 0.0009\%$ in a 500 $\mu$m thick sample, which is of the same order of magnitude as the one in the DSTMS example.

### 5. CONCLUSIONS

We have proposed and analyzed a method to detect coherent radiation by means of diffraction on the induced refractive index grating whose phase is velocity-matched to the one of an easily detectable auxiliary probe wave. The electric field grating that drives the effect may be obtained by a standard two-wave interferometer or by a compact Lloyd-type interferometer. The expected diffracted probe wave is potentially obtained over a black background. Due to the nearly instantaneous electro-optic effect, the response time of the measurement will be limited only by the speed of the conventional detector used to observe the diffraction. Conversely, the technique proposed here may be used for material characterization, for instance for the extraction of material parameters in terahertz time-domain spectroscopy.

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