Probing ultrafast optomagnetism by terahertz Cherenkov radiation

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We put forward Cherenkov-type terahertz emission from a moving pulse of magnetization as a method to explore ultrafast optomagnetic phenomena. We propose to use a structure comprising a slab of transparent magnetooptic material coupled to an output prism. An ultrashort laser pulse propagates in the slab and produces transient magnetization via the inverse Faraday effect. The moving magnetization emits a Cherenkov cone of terahertz waves in the output prism. We developed a theory that predicts the detectability of the radiation for a terbium gallium garnet slab covered with a Si prism.

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I. INTRODUCTION

Ultrafast optical manipulation of magnetization in solids has attracted much interest in recent years as a new fundamental phenomenon in the physics of magnetism with promising potential applications in magnetic storage and information processing technology. The mechanisms of the manipulation can be of thermal or nonthermal origin.[1,2] In thermal effects, such as laser-induced demagnetization[3] and spin reorientation,[4] the magnetic changes result from optical absorption followed by a rapid increase of temperature. Potential applications of thermal effects include the ultrafast magnetization switching in ferromagnetic metal alloys.[5] Nonthermal effects are based on Raman-type nonlinear optical processes, in particular, on the inverse Faraday effect (IFE): the generation of static magnetization by circularly polarized light. Although this effect was predicted as early as 50 years ago[6,7] and observed for relatively long (30 ns) laser pulses soon after the prediction,[8] the ultrafast IFE was demonstrated only very recently.[9] The mechanism of the effect on the subpicosecond time scale is still under debate.[10–13]

A useful tool for investigating the light-induced ultrafast magnetic phenomena is the observation of terahertz radiation from magnetic materials excited by femtosecond laser pulses. This technique, a kind of terahertz emission spectroscopy, has been first used to study fast (thermal) demagnetization induced in ferromagnetic metals (Ni and Fe) by laser pulse irradiation.[14,15] Recently, measuring terahertz emission was used to detect optically excited oscillations of magnetization at the frequency ($\sim$1 THz) of antiferromagnetic resonance in NiO.[16–19] Although the detailed mechanism of the excitation remains unclear, it was speculated that IFE could mediate triggering the oscillations via generation of effective magnetic field acting on the spins.[20] The contradiction of this speculation with the excitation by a linearly polarized laser pulse was attributed to linear magnetic birefringence of NiO.[16,17] The experiments in Refs. 16–19 do not, however, provide direct information about IFE because they are based on measuring the radiation from the magnon oscillations lasting a long time after termination of the laser pulse rather than from the transient magnetization produced via IFE during the action of the pulse. In view of the fundamental role of IFE in femtomagnetism and controversy about its mechanism, a more direct approach to exploring IFE by means of terahertz emission spectroscopy is required.

For direct probing of the ultrafast IFE, both the experimental configuration and the sample material should be entirely different from those in Refs. 16–19. First, the pump laser beam should be focused to a size smaller than the terahertz wavelength (<300 μm), unlike Refs. 16–19 where laser beams with a diameter $\geqslant 1$ mm were used. Indeed, the magnetization induced via IFE is longitudinal (i.e., directed along the laser beam[6,7]) and at a large diameter of the beam it forms a quasiplane longitudinal source which can not radiate a transverse electromagnetic wave. However, as it has been known for optical rectification of femtosecond laser pulses in electro-optic crystals—an electric counterpart of the ultrafast IFE—the longitudinal source (polarization) induced by a focused laser pulse can emit terahertz radiation via the Cherenkov radiation mechanism.[21] Second, to produce a Cherenkov cone large enough for detection, the laser pulse should propagate a long distance in the sample.[21] For this reason, a thick sample of optically transparent material should be used rather than thin ($\leqslant 100$ μm) strongly absorbing (with ~ 100 cm^{-1} absorption coefficient) films.[16–19] Third, the generated Cherenkov cone can suffer total internal reflection at the sample boundary due to a large terahertz refractive index of the sample. To overcome this limitation, a special shaping of samples or prism output coupling should be used.[22,23]

Thus we put forward terahertz Cherenkov radiation as a method to explore ultrafast optomagnetic phenomena. As we rigorously demonstrate below, the transient magnetization induced in a magnetooptic material by an ultrashort laser pulse via IFE emits terahertz Cherenkov radiation. To maximize the radiation, we propose to use a structure consisting of a slab of an optically transparent magnetooptic material and an output prism attached to a lateral surface of the slab (see Fig. 1). The circularly polarized femtosecond laser pulse focused to a line by a cylindrical lens propagates in the slab and produces ultrafast magnetization via IFE. The moving magnetization emits Cherenkov wedge of terahertz waves in the output prism. We develop a theory of Cherenkov radiation in such a structure and apply it to a terbium gallium garnet (TGG) slab covered with high-resistivity Si prism and pumped by a Ti:sapphire laser (800-nm wavelength). TGG has been chosen due to its

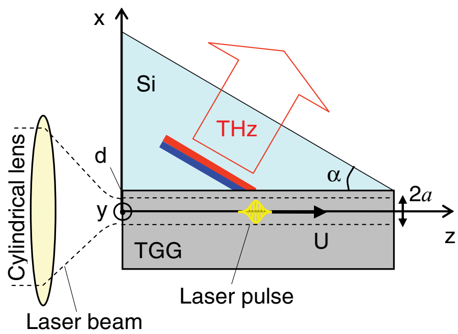

FIG. 1. (Color online) Generation scheme. An optical pulse focused to a line propagates in a slab of magnetooptic material (TGG) and excites Cherenkov wedge of terahertz (THz) radiation in the output (Si) prism.

excellent optical properties and the highest Verdet constant at room temperature. Importantly, in this centrosymmetric cubic crystal, terahertz generation via electrooptic rectification is forbidden.

Probing ultrafast optomagnetism with terahertz Cherenkov radiation can serve as a complimentary tool to the conventional pump-probe technique.[1] An advantage of our scheme over those in Refs. 16–19 is that it allows one to observe directly the radiation from a transient nonlinear magnetization produced via IFE rather than subsequent magnon oscillations triggered by the transient effective magnetic field (if such triggering indeed occurs). In particular, an experimental observation of terahertz Cherenkov radiation in the proposed here scheme would unequivocally prove a genuine ultrafast magnetization.

In classical electrodynamics, Cherenkov radiation from a relativistically moving magnetic dipole has been a subject of discussion for many years.[24–26] It is recognized now that the radiation is different for two types of the dipole: small electric current loop and two magnetic monopoles separated by a small distance. The problem of understanding the nature of this difference is related to the correct modeling of how a moving dipole acts on the medium flowing through the dipole.[25] In this paper, we study Cherenkov radiation from a relativistic magnetic dipole of other, third, type, i.e., from a magnetization created in a magnetooptic material by a propagating laser pulse. Although the magnetized region moves with the group velocity of the optical pulse, there is no moving matter. The magnetization is created by the front edge of the pulse and extinguished by its rear edge. In this practically feasible case, there is not such a problem as for the moving dipole made of matter: the magnetization is consistently defined in the reference frame of the medium.

II. MODEL

The coordinate system is introduced in Fig. 1. In the y direction, the beam width is much greater than the terahertz wavelength (of several millimeters). This allows us to consider the beam as a two-dimensional one with fields independent of y. The laser pulse propagates in the $+z$ direction with the group velocity U. We neglect the optical pulse distortion and write the optical intensity envelope as a given function of $\xi = t - z/U$ and x: $I(\xi,x) = I_0 F(\xi) G(x)$. The peak optical intensity I_0 is $I_0 = c(8\pi)^{-1} n_{\rm opt} E_0^2$, where E_0 is the maximum of the optical field envelope, $n_{\rm opt}$ is the optical refractive index of the slab material, and c is the velocity of light (all formulas in the paper are in cgs units). We use Gaussian functions for the temporal envelope $F(\xi)$ and transverse profile $G(x)$,

$$F(\xi) = \exp(-\xi^2/\tau^2), \quad G(x) = \exp(-x^2/a^2), \tag{1}$$

where τ is the pulse duration [the standard full-width at half maximum (FWHM) is $\tau_{\rm FWHM} = 2\sqrt{\ln 2}\tau \approx 1.7\tau$] and a is the pulse transverse size ($a_{\rm FWHM} = 2\sqrt{\ln 2}a$). We assume that a is small as compared to the distances between the center of the laser path ($x = 0$) and the slab's boundaries (in particular, $a \ll d$) so that the boundaries do not distort the pulse. Therefore the velocity U is defined by the optical group refractive index of the slab material n_g: $U = c/n_g$. The slab is assumed to be sufficiently thick along the x axis (with a thickness >100 μm) to ensure filtering out the terahertz pulses arriving at the prism after reflection from the slab's lower boundary by electro-optic sampling. This allows us to treat the slab as a half space.

The magnetization produced via IFE in a magnetooptic material can be written as[7]

$$\mathbf{M}^{\rm NL} = \pm \mathbf{z} m I(\xi,x), \quad m = V\omega_{\rm opt}^{-1} I_0, \tag{2}$$

where V is the Verdet constant of the material, $\omega_{\rm opt}$ is the optical frequency, and the upper and lower signs are taken for the right and left circularly polarized light, respectively. For linearly polarized light, $\mathbf{M}^{\rm NL} = 0$. According to Eq. (2), $\mathbf{M}^{\rm NL}(\xi,x)$ follows the optical intensity envelope, i.e., it represents a Gaussian pulse moving with the velocity U. Since the magnetization (2) is proportional to the optical intensity, it is quadratically nonlinear with respect to the optical field. This is emphasized by the superscript NL, to distinguish $\mathbf{M}^{\rm NL}$ from the magnetization induced in the material due to its linear magnetic susceptibility.

We note that Eq. (2) has never been proved on the subpicosecond time scale, neither theoretically nor experimentally. Moreover, it was recently argued that theory of IFE[7] should be revisited for the regime of ultrafast magnetization dynamics.[10–12] An experimental observation of terahertz Cherenkov radiation in the proposed here scheme can serve as a probe of Eq. (2).

III. THEORETICAL FORMALISM

To find the terahertz radiation generated by the moving magnetization (2), we use Maxwell's equations with $\mathbf{M}^{\rm NL}$ included as a source. Applying Fourier transform with respect to ξ [ω is the corresponding Fourier variable (frequency); $\tilde{}$ will denote quantities in the Fourier domain] we reduce the equations to the form

$$\nabla_\omega \times \tilde{\mathbf{E}} = -\frac{i\omega}{c}\mu\tilde{\mathbf{H}} - \frac{4\pi i\omega}{c}\tilde{\mathbf{M}}^{\rm NL}, \quad \nabla_\omega \times \tilde{\mathbf{H}} = \frac{i\omega}{c}\varepsilon\tilde{\mathbf{E}}, \tag{3}$$

where the nabla operator ∇_ω has components $(\partial/\partial x, 0, -i\omega U^{-1})$ and the permittivity $\varepsilon(x)$ and permeability $\mu(x)$ in the terahertz range are ε_s and μ_s in the slab ($x < d$) and ε_p and 1 in the prism ($x > d$), respectively. In Eq. (3), $\tilde{\mathbf{M}}^{\rm NL} = \pm \mathbf{z} m \tilde{F}(\omega) G(x)$, with $\tilde{F} = \tau(2\sqrt{\pi})^{-1}\exp(-\omega^2\tau^2/4)$. Projecting Eq. (3) into the coordinate system (see Fig. 1) and eliminating $\tilde{H}_x$ and $\tilde{H}_z$ we obtain an equation for $\tilde{E}_y$ (for the right circularly polarized optical pump):

$$\mu\frac{\partial}{\partial x}\left(\frac{1}{\mu}\frac{\partial \tilde{E}_y}{\partial x}\right)+\kappa^2\tilde{E}_y=-\frac{4\pi\mu\omega^2 m}{c^2}\tilde{F}\frac{\partial}{\partial x}\left(\frac{G}{\mu}\right), \quad (4)$$

where $\kappa^2=(\omega/c)^2[\varepsilon(x)\mu(x)-n_{\rm g}^2]$. We solve Eq. (4) in the regions $x<d$ and $x>d$ and match the solutions by the boundary conditions of continuity of $\tilde{E}_y$ and $\mu^{-1}\partial\tilde{E}_y/\partial x$ that arise after integrating Eq. (4) across the boundary at $x=d$. We arrive at the following expressions for the electric field transform:

$$\tilde{E}_y=\begin{cases}C_1\exp[-i\kappa_{\rm p}(x-d)], & x>d,\\ C_2\exp[i\kappa_{\rm s}(x-d)]+R(x), & x<d,\end{cases} \quad (5)$$

with

$$R(x)=\frac{2\pi\omega m}{c\kappa_{\rm s}}\tilde{F}\int_{-\infty}^{\infty}dx'\frac{\partial G}{\partial x'}\exp(-i\kappa_{\rm s}|x-x'|), \quad (6)$$

$$C_1=\frac{2R(d)}{1+\kappa_{\rm p}\kappa_{\rm s}^{-1}\mu_{\rm s}},\quad C_2=\frac{1-\kappa_{\rm p}\kappa_{\rm s}^{-1}\mu_{\rm s}}{1+\kappa_{\rm p}\kappa_{\rm s}^{-1}\mu_{\rm s}}R(d). \quad (7)$$

The coefficient $\kappa_{\rm p}$ is κ taken with $\varepsilon=\varepsilon_{\rm p}$ and $\mu=1$ and $\kappa_{\rm s}$ is κ taken with $\varepsilon=\varepsilon_{\rm s}$ and $\mu=\mu_{\rm s}$. To ensure the generation of Cherenkov radiation in the prism, the condition $\varepsilon_{\rm p}>n_{\rm g}^2$ should be fulfilled; otherwise $\kappa_{\rm p}$ is purely imaginary and only evanescent fields will be generated in the prism.

In Eq. (5), the terms with $C_{1,2}$ are the free-wave response, i.e., the waves freely propagating from the slab-prism boundary to $x\to\pm\infty$, respectively, and $R(x)$ is the forced-wave response including the near field of the nonlinear source (2) and (for $\varepsilon_{\rm s}>n_{\rm g}^2$) the Cherenkov wedge in the slab. Outside the laser beam, i.e., at $|x|\gg a$, integral (6) can be approximately evaluated as

$$R(x)\approx\mp i2\pi^{3/2}m\omega ac^{-1}\tilde{F}\exp\left(\mp i\kappa_{\rm s}x-\kappa_{\rm s}^2a^2/4\right), \quad (8)$$

where upper and lower signs are for $x\gg a$ and $x\ll -a$, respectively. Since we assumed $d\gg a$, Eq. (8) can be used to define $R(d)$ in Eq. (7).

Applying inverse Fourier transform to Eq. (5) with $R(x)$ and $R(d)$ given by approximate Eq. (8), we obtain for $x<d$:

$$E_y(\xi,x)=\frac{2\pi^{3/2}ma\tau}{c\tau_{\rm eff}}\frac{\partial}{\partial\xi}\left[\mp e^{-\frac{(\xi-\frac{|x|}{c}\sqrt{\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2})^2}{\tau_{\rm eff}^2}}-\frac{1-\kappa_{\rm p}\kappa_{\rm s}^{-1}\mu_{\rm s}}{1+\kappa_{\rm p}\kappa_{\rm s}^{-1}\mu_{\rm s}}e^{-\frac{(\xi+\frac{x-2d}{c}\sqrt{\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2})^2}{\tau_{\rm eff}^2}}\right] \quad (9)$$

[the sign is chosen as in Eq. (8)] and for $x>d$:

$$E_y(\xi,x)=-\frac{4\pi^{3/2}ma\tau}{c\tau_{\rm eff}(1+\kappa_{\rm p}\kappa_{\rm s}^{-1}\mu_{\rm s})}\frac{\partial}{\partial\xi}e^{-\frac{(\xi-\frac{x-d}{c}\sqrt{\varepsilon_{\rm p}-n_{\rm g}^2}-\frac{d}{c}\sqrt{\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2})^2}{\tau_{\rm eff}^2}} \quad (10)$$

with the effective optical pulse duration $\tau_{\rm eff}=[\tau^2+a^2c^{-2}(\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2)]^{1/2}$. In Eq. (9), the first term describes the Cherenkov wedge in the slab and the second term gives the part of the wedge reflected from the slab-prism boundary. Equation (10) describes the Cherenkov wedge transmitted to the prism. The opening angles of the Cherenkov wedge in the slab ($\theta_{\rm s}$) and prism ($\theta_{\rm p}$) are defined by formulas $\cot\theta_{\rm s}=(\varepsilon_{\rm s}\mu_{\rm s}/n_{\rm g}^2-1)^{1/2}$ and $\cot\theta_{\rm p}=(\varepsilon_{\rm p}/n_{\rm g}^2-1)^{1/2}$. According to Eqs. (9) and (10), the field distribution across the Cherenkov wedge is given by the derivative of the optical intensity envelope, i.e., it consists of two adjacent pulses of opposite polarities. The parameter $\tau_{\rm eff}$ in the Gaussian functions in Eqs. (9) and (10) and, therefore, the duration of the pulses at the Cherenkov wedge (the wedge's thickness), depend both on τ and a. For $a\gg c\tau(\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2)^{-1/2}$, $\tau_{\rm eff}$ depends mainly on a, i.e., the field distribution across the wedge becomes smoother with increasing a. For $a\ll c\tau(\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2)^{-1/2}$, $\tau_{\rm eff}$ depends mainly on τ. According to Eq. (10), the maximum value of the electric field at the Cherenkov wedge in the prism is

$$|E_y|_{\max}=\frac{2^{5/2}\pi^{3/2}ma\tau}{c\tau_{\rm eff}^2e^{1/2}\left[1+\mu_{\rm s}\left(\varepsilon_{\rm p}-n_{\rm g}^2\right)^{1/2}\left(\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2\right)^{-1/2}\right]} \quad (11)$$

at $\xi-(x-d)c^{-1}(\varepsilon_{\rm p}-n_{\rm g}^2)^{1/2}-dc^{-1}(\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2)^{1/2}=\pm\tau_{\rm eff}/2^{1/2}$. For a fixed energy of the optical pulse, $ma\tau=$ const in Eq. (11) and $|E_y|_{\max}\propto\tau_{\rm eff}^{-2}$. Thus, for a given τ, a decrease in a leads to an increase in $|E_y|_{\max}$; however, when a becomes smaller than $c\tau(\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2)^{-1/2}$, further focusing adds little to the terahertz field magnitude. Similarly, for a given a, a decrease in τ leads to an increase in $|E_y|_{\max}$ until τ becomes smaller than $ac^{-1}(\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2)^{1/2}$. If we fix the optical intensity rather than energy ($m=$ const) and also fix τ in Eq. (11), then $|E_y|_{\max}\propto a/\tau_{\rm eff}^2$ and an optimal transverse size $a_{\rm opt}=c\tau(\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2)^{-1/2}$ appears that maximizes $|E_y|_{\max}$ to $(2\pi)^{3/2}e^{-1/2}m[(\varepsilon_{\rm s}\mu_{\rm s}-n_{\rm g}^2)^{1/2}+\mu_{\rm s}(\varepsilon_{\rm p}-n_{\rm g}^2)^{1/2}]^{-1}$.

IV. ANALYSIS FOR TGG/Si-PRISM STRUCTURE

Let us now apply the developed theory to a structure consisting of a Si prism and TGG slab pumped by Ti:sapphire laser (800-nm wavelength). For TGG, we use following parameters in the terahertz range: $\mu_{\rm s}\approx 1$ and the refractive index $n_{\rm s}=(\varepsilon_{\rm s}\mu_{\rm s})^{1/2}=3.5-i0.015\cdot\nu$, where ν is the frequency in THz (from our measurement). In the optical range, we use $n_{\rm opt}=1.95$ and $n_{\rm g}=2.14$.[27,28] The Verdet constant of TGG at 800 nm is $V=-87$ rad T^{-1} m^{-1} (or -0.29 min Oe^{-1} cm^{-1}).[28] For Si, the refractive index in the terahertz range is $n_{\rm p}=\varepsilon_{\rm p}^{1/2}=3.418$.[29]

For the given $n_{\rm s}$ and $n_{\rm g}$, the parameter $\tau_{\rm eff}$ can be expressed in terms of practical units as $\tau_{\rm eff}\approx\sqrt{\tau^2+(9.2a)^2}$, where $\tau_{\rm eff}$ and τ are in fs and a is in μm. Equation (11) becomes

$$|E_y|_{\max}\left[\frac{\rm V}{\rm cm}\right]\approx 1.1\times10^7\frac{W_{\rm opt}[{\rm mJ/cm}]}{\tau_{\rm eff}^2[{\rm fs}]}, \quad (12)$$

where $W_{\rm opt}$ is the energy of the pump optical pulse per unit length along the y axis. For $W_{\rm opt}=100$ μJ/cm, $\tau_{\rm FWHM}=100$ fs ($\tau\approx 60$ fs), and $a_{\rm FWHM}=50$ μm ($a\approx 30$ μm), we obtain $\tau_{\rm eff}\approx 282$ fs and $|E_y|_{\max}\approx 14$ V/cm. When exiting from the Si prism, the electric field experiences an enhancement by a factor of $\approx$1.55 and thus the peak field in the free space is $E_v\approx 21$ V/cm. This value is comparable with terahertz emission from a conventional photoconductive antenna.[30] Focusing the terahertz radiation emitted from a TGG/Si-prism structure of 1 cm length (along the z axis) and 1 cm width (along the y axis) to a spot of $\sim$1 mm size

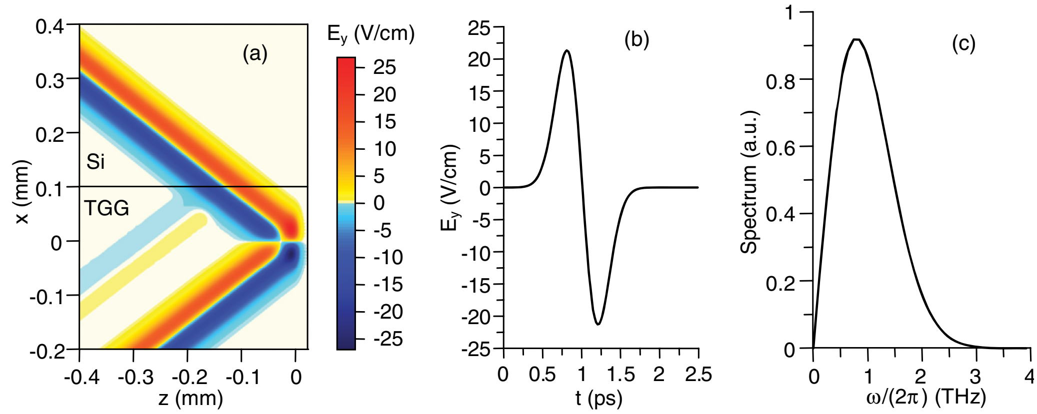

FIG. 2. (Color online) (a) Snapshot of the terahertz field $E_y(x,z,t)$ at $t = 0$ in TGG/Si structure pumped by a Ti:sapphire laser pulse with $\tau_{\rm FWHM} = 100$ fs, $a_{\rm FWHM} = 50$ μm, and $I_0 = 177$ GW/cm^2 ($W_{\rm opt} = 100$ μJ/cm). The pulse propagates at $d = 100$ μm from the TGG-Si interface. (b) Terahertz wave form emitted from the Si prism to the free space. (c) Terahertz amplitude spectrum.

can increase the terahertz field by an order of magnitude. Hopefully, even higher terahertz fields can be achieved by cryogenic cooling of TGG due to an increase in the Verdet constant.[31]

Figure 2(a) shows the distribution of E_y in TGG/Si structure calculated on the basis of Eqs. (5)–(7). Since $n_{\rm p} \approx n_{\rm s}$, the opening angle of the Cherenkov wedge is almost the same in TGG and Si: $\theta_{\rm p} \approx \theta_{\rm s} \approx 39^\circ$. The double wedge structure of the radiation pattern in TGG agrees with Eq. (9). To output efficiently the Cherenkov radiation, the Si prism should be cut with its exit face parallel to the wave front of the radiation, i.e., at $\alpha \approx \theta_{\rm p} \approx 39^\circ$ (see Fig. 1). Figure 2(b) shows the terahertz wave form in the free space for the same parameters as in Fig. 2(a). The wave form comprises two pulses of opposite polarity, in accord with Eq. (10). The peak field agrees with Eq. (12). The corresponding spectrum $|\tilde{E}_y(\omega)|$ is shown in Fig. 2(c). Changing the optical pump polarization from right to left circular will reverse the polarity of the pulses in Fig. 2(b).

Analysis of experimental terahertz wave forms can be used for verification of Eq. (2) on the subpicosecond time scale. First, the dependence of $\mathbf{M}^{\rm NL}$ direction on the pump polarization can be verified from the polarity of the terahertz pulses. Second, if $\mathbf{M}^{\rm NL}$ does not follow the optical intensity envelope, as predicted by Refs. 11,12, it can be detected from the shape of the wave form. Third, measuring the amplitude of the wave form one can derive a value of the Verdet constant in the ultrafast regime.

V. CONCLUSION

To conclude, we have proposed a technique to explore ultrafast optomagnetic phenomena. The technique employs Cherenkov emission of terahertz waves from a moving pulse of laser-induced magnetization in a slab of magnetooptic material covered with an output prism. We developed a theory that predicts for a TGG/Si-prism structure the terahertz yield comparable to that from conventional terahertz sources. In particular, ∼20 V/cm terahertz field (non focused) can be generated in such a structure pumped by 100 fs, 100 μJ pulses of Ti:sapphire laser. The observation of Cherenkov radiation in the proposed scheme would testify the ultrafast IFE.

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