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# Interaction of surface plasmon polaritons in heavily doped GaN microstructures with terahertz radiation

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We present the results of experimental and theoretical studies of the surface plasmon polariton excitations in heavily doped GaN epitaxial layers. Reflection and emission of radiation in the frequency range of 2–20 THz including the Reststrahlen band were investigated for samples with grating etched on the sample surface, as well as for samples with flat surface. The reflectivity spectrum for *p*-polarized radiation measured for the sample with the surface-relief grating demonstrates a set of resonances associated with excitations of different surface plasmon polariton modes. Spectral peculiarities due to the diffraction effect have been also revealed. The characteristic features of the reflectivity spectrum, namely, frequencies, amplitudes, and widths of the resonance dips, are well described theoretically by a modified technique of rigorous coupled-wave analysis of Maxwell equations. The emissivity spectra of the samples were measured under epilayer temperature modulation by pulsed electric field. The emissivity spectrum of the sample with surface-relief grating shows emission peaks in the frequency ranges corresponding to the decay of the surface plasmon polariton modes. Theoretical analysis based on the blackbody-like radiation theory well describes the main peculiarities of the observed THz emission. © 2016 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4943063]

### **I. INTRODUCTION**

Optical phenomena associated with surface waves have attracted significant attention of many researches (see, for review, Refs. 1 and 2). During recent years, there has been a growing interest in the study of surface plasmon polaritons (SPPs). The SPPs are electromagnetic (*EM*) waves with specific properties that enable to concentrate *EM* field into deep sub-wavelength volumes. The remarkable properties of the SPP waves are widely utilized in different opto- and bioelectronic applications, such as nanoscale optical devices,<sup>3</sup> near-field scanning microscopy,<sup>4</sup> highly efficient photovoltaics,<sup>5</sup> plasmonic metamaterial waveguides,<sup>6</sup> and bio-chemical sensing devices.<sup>7</sup> Also, the SPP excitations impact the characteristics of infrared photodetectors<sup>8,9</sup> and the luminescence properties of semiconductor heterostructures.<sup>10</sup>

Most studies of SPP excitations deal with the metallic plasmonic structures and the visible or near-infrared radiation. Due to the relatively low carrier concentrations in semiconductor micro- and nanostructures, the typical frequencies of the SPP excitations belong to the mid-infrared and terahertz (THz) frequency ranges. Particularly, in doped semiconductors, the resonance frequencies of SPPs can easily be tuned to the THz frequency range by changing the doping level. The studies of SPPs in these frequency ranges have received less attention. One of the first experimental studies of SPPs in the THz frequency range under external illumination was performed for a *n*-InSb plasmonic structure with a surface grating.<sup>11</sup> For doped samples with electron concentration  $n = 7 \times 10^{18}$  cm<sup>-3</sup>, the sharp dips associated with SPP excitation were experimentally observed in the reflectivity spectra in the range of 6–24 THz. Later on, similar investigations were performed for different kinds of metal/semiconductor plasmonic structures with the use of various experimental techniques. For example, the SPPs in Au grating on GaAs surface<sup>12</sup> or in Si hole arrays<sup>13,14</sup> were studied using THz timedomain spectroscopy. In the last paper, the authors reported the enhancement of THz radiation transmission which was associated with the tunneling effect of the SPPs. Terahertz SPP propagation through gratings structured on silicon surfaces was investigated in Ref. 15.

The narrow-band thermal radiation in mid-IR region was reported for triple layer structure (perforated Ag/SiO<sub>2</sub>/Ag) on Si substrate,<sup>16</sup> for Au grating on Si substrate,<sup>17</sup> and for conducting amorphous carbon composite.<sup>18</sup> Recently, emission of THz radiation under pulsed optical pumping was obtained for graphene on a discontinuous layer of Au.<sup>19</sup> The continuous-wave THz luminescence under electric current pumping was investigated for degenerate *n*-InN epitaxial layers with a *random* grating formed by topographical defects.<sup>20</sup> Together with experimental evidences of the effects of the SPPs, there are several papers which theoretically predict and quantitatively describe the thermal emission properties of the structures with surface gratings.<sup>21,22</sup> THz emission from quantum

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well structures due to 2D plasmon–THz radiation coupling was reported in Refs. 23–25. In the mentioned papers, the observed features of the THz radiation emission were attributed to 2D plasmon scattering on the grating structure.

The frequency selective properties of semiconductor plasmonic structures in the THz frequency range may find a number of applications, particularly, in the fields of THz biosensing and THz bio-imaging. Among different kinds of the semiconductor compounds that are considered as a platform for sensor devices, gallium nitride is assumed to be one of the most favorable. Indeed, the GaN-based structures have unique chemical resistance and are non-toxic. These both properties are important for bio-sensing technologies.

This paper is aimed to provide experimental and theoretical analysis of SPP-EM wave interaction in heavily doped GaN-based microstructures with regular surface-relief grating. The experimental study of the electroluminescence properties of such structure was partially reported in the paper.<sup>26</sup> The paper is organized as follows. In Sec. II, the samples and experimental technique are described. Two experiments were carried out: (i) study of reflectivity spectra of the epitaxial layers for polarized THz radiation, and (ii) study of THz radiation emission spectra from the epitaxial layers under epilayer temperature modulation by pulsed electric field. In Sec. III, we present a theoretical model describing the optical properties (reflectivity and absorptivity) of semi-infinite semiconductor with a regular grating on its surface. The model is based on a rigorous electrodynamic solution of the Maxwell's equations. In Sec. IV, we discuss the experimental data and results of the theoretical simulation. The main results are summarized in Sec. V.

## II. SAMPLES AND EXPERIMENTAL TECHNIQUE

Wurtzite gallium nitride epitaxial layers of 6.2  $\mu$ m thickness were grown by metal-organic vapor phase epitaxy (MOVPE) on *c*-plane sapphire substrates covered by a 2  $\mu$ m undoped GaN buffer layer. The GaN epitaxial layers were heavily doped with silicon donor impurities. The free electron concentration  $n_e = 3.6 \times 10^{19}$  cm<sup>-3</sup> and the mobility  $\mu_e = 122$  cm<sup>2</sup>/V s were determined from Hall measurements at room temperature. In gallium nitride at such high doping levels, an impurity band is formed which overlaps with the conduction band.<sup>27,28</sup> This provides the constant free electron concentration in a wide temperature range from 4.2 to 300 K and higher.

For the purposes of electroluminescent measurements, two Ti/Au electrical contacts of 4 mm length were patterned of 3 mm apart on the top surface of the samples by a standard photolithography process. The epitaxial layer temperature was modulated by applying pulsed voltage to the contacts. A regular grating with a period of  $a_g = 86 \,\mu\text{m}$  was etched on the surface of the samples in the area between the electrical contacts by inductively coupled plasma deep reactive ion etcher (ICP-RIE) using Cl-chemistry. The grating etch depth  $h_g$  was equal to 4.5  $\mu$ m. The ratio of a ridge width  $w_g$  and the grating period  $a_g$  was 1:2. The reference sample with a planar GaN surface was additionally fabricated and studied.

The reflectivity spectra were investigated at room temperature using a Fourier spectrometer Bruker Vertex 80v operating in a rapid-scan mode. The measurement pressure was 4.5 mbar. A mercury lamp and a globar were used as sources of THz radiation. A Mylar beamsplitter was applied. The sample surface was illuminated in the area between the contacts. The beam spot diameter was about 3 mm that is much greater than the grating period. The converging incident radiation beam had an angular aperture of  $\Delta \theta \simeq 16^{\circ}$ . The axis of the beam formed the angle  $\theta = 11^{\circ}$  with the normal to the sample surface. The plane of incidence was oriented perpendicular to the grooves of grating. The *s*- and *p*polarized incident radiations were formed by a grid polarizer on polyethylene. The reflected THz radiation was collected by DLaTGS pyroelectric detector D201/B. An aluminium mirror was used as a reflection reference.

The THz radiation emission from microstructures was measured under epilayer temperature modulation by the pulsed electric field. The sample under investigation was placed in optical closed cycle cryostat (Janis PTCM-4-7) and cooled to the temperature of about 9K. The voltage pulses with duration of about  $2\,\mu s$  and repetition frequency of 87 Hz were applied to the sample contacts. The THz radiation was collected in the direction perpendicular to the sample surface ( $\theta = 0^{\circ}$ ), the angular aperture was  $\Delta\theta \approx 16^{\circ}$  (solid angle  $\Omega = 0.043$  sr). The emission spectra were investigated using the Fourier spectrometer operating in a step-scan mode. A liquid-helium-cooled silicon bolometer (IRLabs) was used as a detector. The signal from the detector was supplied to a low-noise preamplifier (Stanford Research Systems, Model SR560), then the amplified signal was supplied to a gated integrator and boxcar averager (Stanford Research Systems, Model SR250). It should be noted that the duration of the bolometer photoresponse pulse exceeded the duration of the voltage pulse. During the voltage pulse, the epitaxial layer temperature increases due to Joule heating. Then, the epilayer temperature decreases with a time constant of about a few  $\mu$ s due to heat transfer into sapphire substrate, and finally the whole microstructure cools with time constant of about 100  $\mu$ s before the next voltage pulse. We observed experimentally the time evolution of the THz emission intensity by means of a rapid Ge:Ga THz detector with the sensitivity range from 10 to 21 meV. Silicon bolometer which we used for spectral studies of the THz emission was rather slow (maximal operating frequency  $\sim 200 \text{ Hz}$ ), and it broadened the photoresponse pulse in comparison with Ge:Ga detector. That is why it was possible to use rather long gate pulse (15  $\mu$ s) in the gated integrator. Gate pulse was located close to the photoresponse pulse maximum. In this way, we detected the modulation signal which corresponds to the THz emission intensity increase under epilayer heating by pulsed voltage. To increase the accuracy of the measurements, 300 photoresponse pulses were averaged at every position of the scanning mirror in the Fourier spectrometer.

### III. THEORETICAL MODEL OF THE INTERACTION OF PLANE ELECTROMAGNETIC WAVE WITH SURFACE-RELIEF GRATING STRUCTURE

It is well known that the SPP waves are spatially localized excitations that can propagate at the interface between a dielectric and a conducting medium if the following conditions are satisfied:<sup>2</sup>

$$\operatorname{Re} \varepsilon_{c} < 0, \quad \operatorname{Re} \varepsilon_{d} > 0, \quad |\operatorname{Re} \varepsilon_{c}| > |\operatorname{Re} \varepsilon_{d}|, \qquad (1)$$

where  $\varepsilon_c$  and  $\varepsilon_d$  denote dielectric permittivity of the conductor and the dielectric, respectively. In the most simple case of the planar interface between two semi-infinite media, the dispersion law of the SPPs is given by the following expression:<sup>1</sup>

$$k_{\rm SPP}^2 = \left(\frac{\omega}{c}\right)^2 \frac{\varepsilon_{\rm c}(\omega)\varepsilon_{\rm d}(\omega)}{\varepsilon_{\rm c}(\omega) + \varepsilon_{\rm d}(\omega)},\tag{2}$$

where  $k_{\text{SPP}}$  is the longitudinal (in respect to propagation direction) component of the SPP wavevector,  $\omega$  is the frequency, and *c* is the light velocity in vacuum.

In general, for lossy conductor, the dielectric permittivity  $\varepsilon_c$  is a complex function with a nonzero imaginary part. Consequently, Eq. (2) cannot be resolved at real values of  $k_{\text{SPP}}$  and  $\omega$ . The dispersion law of the SPPs can be found, for example, assuming that  $\omega$  is the complex quantity while  $k_{\text{SPP}}$ is the real quantity. In this case, the real and imaginary parts of the frequency correspond to the eigen frequency and decrement of SPP oscillations, respectively. This particular choice can be applied for the characterization of the spectral features of the reflectivity, transmittivity, and absorptivity which are measured at the uniform external illumination (see Ref. 29). These conditions are realized in our experiments (see above Sec. II).

The direct excitation of the SPPs by external *EM* field at the uniform interface is forbidden, because the SPP wavevector is greater than the wavevector of the incident radiation,  $k_{\rm Ph} = \omega \sqrt{\epsilon_{\rm d}(\omega)}/c$ , at a given frequency, and it is impossible to satisfy energy and momentum conservation laws simultaneously. The excitation becomes allowed when spatial inhomogeneity is introduced at the conductor-dielectric interface. For instance, the surface-relief grating can provide a coupling between the incident radiation and the SPP excitations. In the optical experiments, SPP-photon interaction manifests itself as the emergence of the resonant dips/ peaks in the reflection/absorption spectra. Spectral position of these resonances can be estimated using the dispersion law of the SPPs and phase-matching condition<sup>1</sup>

$$k_{\rm SPP} = k_{\rm Ph} \sin \theta + M k_{\rm G},\tag{3}$$

where  $\theta$  is the incident angle,  $\vec{k}_{G}$  is the reciprocal vector of the grating,  $k_{G} = |\vec{k}_{G}| = 2\pi/a_{g}$  (see Fig. 1). The integer numbers  $M = \pm 1, \pm 2, \pm 3,...$ , correspond to different modes of the SPP resonances. The width of the resonance peaks can be provided by the decrement of the SPP oscillations.

Besides the SPP excitation by the external EM wave (see Fig. 1(a)), the backward transformation of the SPPs into outgoing EM wave under SPP scattering on the grating (Fig. 1(b)) can occur. If the former process can be recognized by means of the reflectivity/transmittivity spectra, then the latter one can be detected via EM radiation emission.

Eqs. (2) and (3) predict the spectral positions and widths of the dips/peaks in the reflectivity/absorptivity spectra but



FIG. 1. Illustration of interaction between the SPPs and the *EM* waves at the dielectric–conductor interface with surface-relief grating. (a) Excitation of the SPPs under incidence of *EM* wave at the conductor surface and (b) transformation of the SPPs into *EM* wave under SPP scattering on the surface grating. The gray color shows the localization area of SPPs. The figures correspond to M = +1.

they do not describe their amplitudes and general behavior of the measured spectra. It can be done in the framework of rigorous electrodynamic analysis of the structure with surface-relief grating. Here, we briefly present the theoretical model used in the calculations of the reflectivity/absorptivity spectra of the experimental samples.

Fig. 2 shows the geometry of the modeling structure with the surface-relief grating. The structure is assumed to be uniform along the *y*-direction, and periodical and infinite along the *x*-direction. The latter assumption is reasonable if the size of beam spot is much greater than the grating period.

It is assumed that the plane *EM* wave with a frequency  $\omega$  and wavevector  $\vec{k}_{Ph}\{k_0 \sin \theta, 0, k_0 \cos \theta\}$  uniformly illuminates the structure,  $k_0 = \omega/c$ . A time dependence of the *EM* wave is chosen in the form of  $\exp(-i\omega t)$ . The reflection/ absorption of the *EM* wave can be found by solving the Maxwell equations

$$\operatorname{rot}\vec{H} = -ik_0\varepsilon^*(x,z)\vec{E}, \quad \operatorname{rot}\vec{E} = ik_0\vec{H},$$

with the complex spatially dependent dielectric permittivity,  $\varepsilon^*(x, z)$ . Since in our case  $\varepsilon^*(x, z)$  is the discontinuous function, it is convenient to divide all space to three different regions.



FIG. 2. Geometry of the surface-relief grating structure.

The regions I (z < 0) and III ( $z > h_g$ ) correspond to the half-spaces uniformly filled up by the air (vacuum) and a semiconductor with the dielectric permittivities  $\varepsilon^*(x, z) \equiv \varepsilon_d$ = 1 and  $\varepsilon^*(x, z) \equiv \varepsilon_c$ , respectively. It is assumed that the  $\varepsilon_c$  is a scalar and consists of the two contributions from the lattice and electron subsystems

$$\varepsilon_c = \varepsilon_\omega + \frac{4\pi i \sigma_\omega}{\omega}.\tag{4}$$

The frequency dispersion of the lattice dielectric permittivity  $\varepsilon_{\omega}$  is chosen as for polar semiconductor<sup>30</sup>

$$\varepsilon_{\omega} = \varepsilon_{\infty} + \frac{\left(\varepsilon_0 - \varepsilon_{\infty}\right)\omega_{TO}^2}{\omega_{TO}^2 - \omega^2 - i\gamma\,\omega},\tag{5}$$

where  $\varepsilon_0$  and  $\varepsilon_{\infty}$  are the low frequency and high frequency permittivities, respectively,  $\omega_{TO}$  is the frequency of transverse optical (*TO*) vibrations, and  $\gamma$  is the optical phonon damping. The high-frequency conductivity  $\sigma_{\omega}$  of the electron gas is described by the Drude-Lorentz model, i.e.,

$$\sigma_{\omega} = \frac{e n_e \mu_e}{1 - i \omega m^* \mu_e / e},\tag{6}$$

where  $n_e$  and  $\mu_e$  are the electron concentration and mobility, respectively, *e* and *m*<sup>\*</sup> are the elementary charge and electron effective mass.

In region II  $(0 < z < h_g)$ ,  $\varepsilon^*(x, z)$  is the periodical function with respect to the *x*-coordinate. For one period,  $0 < x < a_g$ ,  $\varepsilon^*(x, z)$  can be written as  $\varepsilon^*(x, z) \equiv \varepsilon_{II} = \varepsilon_c \Theta(w_g - x) + \varepsilon_d \Theta(x - w_g)$ , where  $\Theta(x)$  is the Heaviside step-like function.

The numerical solution of the Maxwell equations was performed based on modified technique of rigorous coupledwave analysis (RCWA).<sup>31–33</sup> The mathematical procedure of RCWA applies the Fourier expansion of the components of EM wave. Using the continuity conditions of tangential components of the *EM* wave at the I–II interface (plane z = 0) and the II–III interface (plane  $z = h_g$ ), the Maxwell equations were reduced to the system of the algebraic equations containing the Fourier coefficients of the appropriate components of reflected and transmitted waves. For the case of p-polarized incident radiation, the system of the algebraic equations is formulated for the Fourier coefficients of the y-component of magnetic fields  $H_{y,m}^R$  and  $H_{y,m}^T$ , where the upper cases R and T correspond to reflected wave into region I and transmitted wave into region III, respectively. For the case of s-polarized incident radiation, the system contains Fourier coefficients of the y-component of electric field  $E_{y,m}^R$  and  $E_{y,m}^T$ . The details of the mathematical procedure are given in the Appendix.

Numerical solution of the discussed system (A13) allows us to obtain the distribution of *EM* field in the whole space, including the near-field zone and calculate the reflectivity  $R_m$  for the any *m*-th diffraction order (diffraction efficiency). For *p*-polarization,  $R_m$  reads

$$R_m = \operatorname{Re}\left[\frac{\sqrt{\varepsilon_d k_0^2 - \beta_m^2}}{k_0 \cos \theta}\right] |H_{y,m}^R|^2, \tag{7}$$

where  $\beta_m$  is given by Eq. (A3). For *s*-polarization,  $R_m$  is given by the same formula (7) with the substitution  $H^R_{y,m} \to E^R_{y,m}$ .

Below, we will present the results of the calculations of the zero-order diffraction efficiency,  $R_0(\omega, \theta) = |H_{y,0}^R|^2$ , total diffraction efficiency,  $R_{tot}(\omega, \theta) = \sum_m R_m$ , and absorptivity,  $A(\omega, \theta) = 1 - R_{tot}(\omega, \theta)$ . Note that these quantities are the functions of the both frequency and incident angle. The developed theory is applied for quantitative description of the reflectivity/emissivity spectra and comparison with the measured characteristics of gallium nitride sample with the surface-relief grating.

# IV. EXPERIMENTAL RESULTS, THEIR ANALYSIS, AND DISCUSSION

#### A. Reflection from the GaN layer with planar surface

We start with the study of the reflectivity spectra of the GaN epitaxial layer with planar surface (reference samples). Below, we will denote the reflectivity and the absorptivity of the reference sample as R' and A', respectively. The data obtained for the reference sample help us afterwards to recognize the peculiarities related to SPP excitations in the epitaxial layers with the surface-relief grating. The measurements of the reflectivity were carried out for the wide spectral range 10–85 meV (2.5–20 THz), that particularly includes the Reststrahlen band of the GaN.

Generally, the wurtzite GaN epitaxial layer grown on c-plane sapphire substrate is optically uniaxial crystal with the optical axis parallel to the growth direction, and its crystal lattice is characterized by two main permittivity tensor components,  $\varepsilon_c^{\parallel}$  (for  $\vec{E} \parallel c$ ) and  $\varepsilon_c^{\perp}$  (for  $\vec{E} \perp c$ ), both values are presented in Table I. But in accordance with our estimates for small incidence angles ( $\theta \sim 11^\circ$ ), the anisotropy can be neglected in the simulation of the reflectivity and absorptivity spectra. At the theoretical analysis of the optical properties of the epitaxial GaN layers with planar and profiled surfaces, we use the isotropic approximation and the scalar complex dielectric permittivity given by Eqs. (4)–(6) with lattice parameters corresponding to the case of  $\vec{E} \perp c$ .

The results of the measurements and calculations of the reflectivity spectra for the reference sample are shown in Fig. 3(a). As seen, the features of the experimental spectra (thick lines) are well described by the theoretical curves (thin lines) obtained in accordance with analytical formulas (see Ref. 39) adopted for the epitaxial *n*-doped layer of a finite thickness. In the calculations, four parameters of

TABLE I. Model parameters for optical phonons in wurtzite GaN (data are taken from Refs. 34–38).

$\overline{\epsilon_{\infty}^{\perp}}$	5.4
$\varepsilon_0^{\perp}$	9.5
$\hbar\omega_{TO}^{\perp}$	69.3 meV
$\gamma^{\perp}$	$7.5  imes 10^{11}  s^{-1}$
$\varepsilon_{\infty}^{\parallel}$	5.5
$\varepsilon_0^{\parallel}$	10.5
$\hbar \omega_{TO}^{\parallel}$	66 meV
$\gamma^{\parallel}$	$11 \times 10^{11}  \mathrm{s}^{-1}$



FIG. 3. (a) Reflectivity spectra for *p*- and *s*-polarized radiation for the GaN epitaxial layer with planar surface. The incidence angle  $\theta = 11^{\circ}$ . The thick lines present the experimental data (spectral resolution is 0.25 meV), thin lines depict the simulation results. Dash-dotted and dash-dot-dotted lines are the reflectivity spectra for *p*- and *s*-polarized radiation, respectively, calculated for semi-infinite GaN with  $n_e = 10^{17}$  cm<sup>-3</sup> and the same other parameters. Here and below  $f = \omega/2\pi$ . (b) Frequency dispersion of the real and imaginary parts of the conductor permittivity  $\varepsilon_c$ . The calculation was made according to Eqs. (4)–(6). Model parameters are the same as in the reflectivity spectra calculation.

wurtzite GaN were fixed at the following values:  $\varepsilon_0 = 9.5$ ,  $\varepsilon_{\infty} = 5.4$  and  $\hbar \omega_{TO} = 69.3$  meV (see Table I), and  $m^* = 0.2$  $m_0$  (see Ref. 40). Three other model parameters  $n_e$ ,  $\mu_e$ , and  $\gamma$  were obtained by fitting. The best fit was given by  $n_e$  $= 1.9 \times 10^{19} \text{ cm}^{-3}$ ,  $\mu_e = 179 \text{ cm}^2/\text{V}$  s, and  $\gamma = 7.5 \times 10^{11} \text{ s}^{-1}$ . In Fig. 3(b), the *n*-GaN dielectric permittivity calculated at the same parameters is shown. Note that the real part of the permittivity is negative at  $\hbar \omega < 62 \text{ meV}$ . It is in this spectral range that we observed peculiarities related to SPPs in the reflection and emission (see below Subsections IVB and **IV C**). Constant of the optical phonon damping  $\gamma$  depends on the crystal lattice quality and has been reported to differ in a wide range from  $6.6 \times 10^{11} \text{ s}^{-1}$  to  $1.9 \times 10^{12} \text{ s}^{-1}$  (Refs. 41 and 42). The value for  $\gamma$  determined from our optical measurements lies in the lower end of reported values and indicates high crystal quality. It should be noted that the values of the electron concentration and mobility obtained from reflectivity spectra differ slightly from the values determined by Hall measurements. The latter are related to the average values across the whole epilayer, but the former characterize only a subsurface region of the epilayer. The optically determined values are used in the further analysis of the profiled epitaxial layers.

Layers thicker than  $1 \,\mu m$  with very high electron concentrations  $(n_e > 10^{19} \text{ cm}^{-3})$  were found opaque in the measured spectral range. Additionally, the numerical procedure developed for semi-infinite media described in the Appendix was verified to yield the same theoretical spectra in the limit of  $h_g \rightarrow 0$ . As seen in Fig. 3(a), the reflectivity spectra have two distinctive behaviors depending on the ratio between the contribution of electron and optical phonon subsystems to the complex permittivity. In the lower frequency range (f < 10 THz,  $\hbar \omega < 40$  meV), the electron contribution dominates resulting in weak frequency dependence of the reflectivity. Such behavior corresponds to the Drude-Lorentz model (Eq. (6)) with strong electron scattering,  $\omega m^* \mu_e/e < 1$ . The strong frequency dependence of the reflectivity is observed in the spectral range of 10-20 THz where the contribution of the phonon subsystem dominates over electrons. The significant decrease of the reflectivity (up to  $R \simeq 0.4$ ) and corresponding increase of the absorptivity (up to 0.6) are observed at the photon energy close to TO-phonon resonance (at  $\omega \simeq 0.9 \times \omega_{TO}$ ). Also, due to the small incident angle, both experiment and theory give almost the same results for the *s*- and *p*-polarizations.

It should be noted that the obtained reflectivity spectra in the *Reststrahlen* band (in the spectral range between  $\omega_{TO}$ and  $\omega_{LQ} = 1.33 \times \omega_{TQ}$ ) have unusual shape. Typically, in this spectral range, the reflectivity exceeds 0.8, and there is a wide spectral interval outside of *Reststrahlen* band where the reflectivity is less than 0.3. Such spectra were observed in *n*-GaN with rather low doping level of  $\leq 10^{17}$  cm<sup>-3</sup> (see, for instance, Refs. 42 and 43). As an illustration, the reflectivity spectra calculated for semi-infinite GaN with electron concentration  $10^{17}$  cm<sup>-3</sup> are shown in Fig. 3(a) by the dasheddotted and dashed-dotted-dotted lines. The samples in this study have essentially higher doping level, and thus an inequality,  $\omega_p > \omega_{LO}$ ,  $\omega_{TO}$ , between the frequencies of the bulk plasmon,  $\omega_p = \sqrt{4\pi n_e e^2/\varepsilon_{\infty} m^*}$ , and optical phonons occurs. At assumed GaN parameters,  $\hbar \omega_p = 156 \text{ meV}$ (f = 38 THz). Subsequently, at  $\omega \le \omega_{LO}$ , the superposition of the Reststrahlen band and the plasma reflection band takes place, and the reflectivity exceeds 0.8 except of the narrow dip corresponding to the low-frequency plasmon-phonon mode located closely to  $\omega_{TO}$ . But even in this dip, the reflectivity exceeds 0.4.

# B. Reflection from the GaN layer with surface-relief grating

Experimental reflectivity spectra of the epitaxial layer with a surface-relief grating are presented in the main panels of Figs. 4(a) and 4(b) for *p*- and *s*-polarized radiation, respectively. As seen in the figures, these spectra differ cardinally from the spectra of the planar epitaxial layer (Fig. 3(a)) and have several specific features.



FIG. 4. Reflectivity spectra for the profiled epitaxial layer for *p*-polarized (a) and for *s*-polarized (b) radiation. Thick lines present the experimental data (spectral resolution is 0.25 meV), the thin lines depict the simulation results for the angle-averaged reflectivity of the zero diffraction order  $\langle R_0 \rangle_0$ . The inset in panel (a): simulation results of reflectivity of the zero diffraction order  $R_0$  at  $\theta = 11^\circ$ . The inset in panel (b): angular distribution of the radiation intensity in the incident beam. The arrows denote the resonance frequencies corresponding to the phase-matching condition (Eq. (3)) and the SPP dispersion law (Eq. (2)).

The first feature is associated with an excitation of SPPs in the profiled epitaxial layer. The experimental spectrum for p-polarization demonstrates a sequence of dips, whereas the spectrum for *s*-polarization does not contain similar dips. Actually, it is known that only EM radiation having a component of electric field vector perpendicular to the grating grooves can excite surface waves.<sup>44</sup> This causes dissipation of EM wave energy and is seen as a decrease of reflectivity. The excitation of SPPs has a resonant behavior, and the resonant frequencies can be found using the phase-matching condition and the SPP dispersion law (see Eqs. (2) and (3)). It is seen from Fig. 4(a) that the calculated resonant frequencies corresponding to the phase-matching condition at  $M = \pm 1, +2,$ and -3 match well the dip positions in the experimental reflectivity spectrum. Note that the observed resonant peculiarities of the reflectivity spectrum are a manifestation of the Wood's anomaly<sup>45</sup> in THz frequency range.

The second feature seen in Figs. 4(a) and 4(b) is the collapse of the Reststrahlen band that is well pronounced in the sample with planar surface. Apparently, this effect is associated with the measurement conditions. The detailed explanation is given below.

For quantitative explanations of the obtained experimental results, we performed theoretical calculations of the reflectivity, using numerical procedure described in the Appendix. The inset in Fig. 4(a) shows calculated spectrum of the reflectivity  $R_0$  for the zero diffraction order and p-polarized radiation. Here, we use the assumption that the structure is illuminated uniformly by the parallel beam with incident angle  $\theta = 11^{\circ}$ . The spectrum of  $R_0$  shows the very narrow and extremely deep resonances,  $R_0 \approx 0.21$  and  $R_0 \approx 0.23$ , at frequencies 2.92 THz ( $\hbar \omega = 12.1 \text{ meV}$ ) and 4.28 THz ( $\hbar\omega = 17.7$  meV), corresponding to the excitation of the SPP modes for M = -1 and +1, respectively. The estimations of the eigen frequencies of the SPP mode using Eqs. (2) and (3) give almost the same resonance frequencies, at this, the estimated decrements of the corresponding SPP oscillations are equal to -0.0043 THz and -0.0096 THz (respective quality factors for these resonances are 670 and 440). The predicted very high quality factor of the resonances is due to the high electron concentration,  $n_e = 1.9$  $\times 10^{19}$  cm<sup>-3</sup>. At this concentration, the dispersion law of the SPPs is almost linear in considered frequency range and close to the dispersion law of the photons in vacuum,  $\omega = ck$ . Additionally, the electromagnetic field of the SPP modes is weakly localized in the dielectric and penetrate poorly into the conductive semiconductor that stipulates the very low Joule dissipation.

However, compared with simulations, the measured SPP resonances are suppressed and broadened (Fig. 4(a) and inset). This is due to the rather large angular aperture of the incident beam ( $\Delta\theta \simeq 16^{\circ}$ ). Assuming Gaussian form of the angular distribution of the intensity in the incident beam  $I_0(\theta)$  (see the inset in Fig. 4(b)), we calculated angle-averaged reflectivity of the zero diffraction order,  $\langle R_0 \rangle_{\theta} = \int_{\theta_{min}}^{\theta_{max}} d\theta \cos(\theta) I_0(\theta) R_0(\theta) / \int_{\theta_{min}}^{\theta_{max}} d\theta \cos(\theta) I_0(\theta)$ , where  $\theta_{min} = \theta - \Delta\theta/2 = 3^{\circ}$  and  $\theta_{max} = \theta + \Delta\theta/2 = 19^{\circ}$ .

In Fig. 4(a), one can see that the simulated spectrum of  $\langle R_0 \rangle_{\theta}$  for *p*-polarized radiation matches well with the experimental spectrum in the spectral range of 0–10 THz. The frequencies, amplitudes, and widths of the resonance dips in the simulated and experimental spectra are close to each other. The SPP resonances are clearly seen at frequencies of 2.9 THz, 4.4 THz, 8.3 THz, and 8.7 THz. According to Eqs. (2) and (3), we identified that these frequencies correspond to the SPP resonances of orders M = -1, +1, +2, -3, respectively. For these resonances, we analyzed the *EM* fields in the near-field zone (see Fig. 5). In Fig. 5, the Fourier amplitudes  $|H_{y,m}|^2$  for the resonances are shown at z = 0.

At the resonance frequencies 2.9 THz and 4.4 THz  $(M = \pm 1)$ , the Fourier amplitudes  $|H_{y,\pm 1}|^2$  are two orders of magnitude greater than the other ones. This means that for corresponding resonances almost single-mode excitations of the SPPs occur. At two close frequencies 8.3 THz and 8.7 THz (M = +2, -3), the configurations of the near field represent superpositions of different Fourier components,



FIG. 5. Fourier amplitude spectrum of the magnetic field  $H_y$  calculated at z = 0 and  $\theta = 11^{\circ}$ .

i.e., several SPP modes are excited, though the resonant Fourier amplitudes  $|H_{y,+2}|^2$  and  $|H_{y,-3}|^2$  dominate.

In the same spectral range, for *s*-polarized radiation, the calculations do not predict any peculiarities, which is in accordance with the observed spectrum (Fig. 4(b)).

A deviation between the simulated spectrum of the  $\langle R_0 \rangle_{\theta}$ and the observed reflectivity spectrum arises at higher frequencies (f > 10 THz). The deviation increases with increasing the frequency and is observed for the both polarizations. Apparently, this discrepancy is due to the diffraction phenomenon. Indeed, together with the excitation of SPP modes, the surface grating splits the incident beam into several reflected beams, which propagates within different solid angles. According to the well-known grating equations,<sup>46</sup> the angles between the zero order and higher order diffraction beams decrease with increasing frequency. Consequently, in the high frequency range, the angular aperture of detection system (in our case 16°) can partially catch the beams of higher diffraction orders. The estimates show that the beam of the 1st diffraction order is partially caught at f > 10 THz. This explains why observed reflectivity exceeds the angleaveraged reflectivity of the zero diffraction order  $\langle R_0 \rangle_{\theta}$  in this spectral range (see Figs. 4(a) and 4(b)).

As an additional argument of this explanation, we present in Fig. 6 the simulated spectra of the total diffraction efficiency  $R_{tot}$  for both polarizations. In accordance with definition in Sec. III,  $R_{tot}$  includes contributions of all diffraction orders. As expected, the values of  $R_{tot}$  exceed the values of experimental reflectivity R in the spectral range 10–20 THz for the both polarizations (see Figs. 4 and 6). For example, for *p*-polarization at f = 14.5 THz,  $R_{tot} = 0.53$  and R = 0.15.

Remarkably, for *s*-polarized radiation, the total diffraction efficiency for profiled surface  $R_{tot}$  almost coincides with the reflectivity of plane surface R' in the whole considered spectral range (Fig. 6(b)). Particularly, it means that the surface grating does not affect on the absorptivity of *s*-polarized THz radiation ( $A = 1 - R_{tot} \approx A' = 1 - R'$ ). On the contrary, for *p*-polarized THz radiation, the spectral dependencies of  $R_{tot}$  and R' differ significantly (see Fig. 6(a)) at the frequencies corresponding to the excitations of SPP modes. The values of



FIG. 6. Simulated spectra of zero-order ( $R_0$ ) and total ( $R_{tot}$ ) diffraction efficiencies for GaN layer with profiled surface for *p*-polarized (a) and *s*-polarized (b) radiation. The dashed lines depict the simulated reflectivity spectra of the GaN layer with planar surface (R'). The incidence angle  $\theta = 11^{\circ}$ .

 $R_{tot}$  are significantly less than the values of R'. Consequently, the absorptivity spectra for profiled surface  $(A = 1 - R_{tot})$  should demonstrate corresponding absorption peaks. This point will be discussed in detail in Subsection IV C.

To finalize this subsection, let us consider a factor of the optical anisotropy of the wurtzite GaN, which was not taken into account in our model. The anisotropy slightly changes the SPP dispersion and position of the resonances. To account the anisotropy, one should use the more general expression for  $k_{\text{SPP}}^{47}$ 

$$k_{\rm SPP}^2 = \left(\frac{\omega}{c}\right)^2 \frac{\varepsilon_{\rm c}^{\parallel}(\omega)\varepsilon_{\rm d}(\omega)\left(\varepsilon_{\rm d}(\omega) - \varepsilon_{\rm c}^{\perp}(\omega)\right)}{\varepsilon_{\rm d}^2(\omega) - \varepsilon_{\rm c}^{\perp}(\omega)\varepsilon_{\rm c}^{\parallel}(\omega)}.$$
 (8)

Analytical expressions for  $\varepsilon_{c}^{\parallel}(\omega)$  and  $\varepsilon_{c}^{\perp}(\omega)$  are given by the relations similar to Eqs. (4) and (5) where all parameters of the crystal lattice are marked by the superscripts " $\parallel$ " and " $\perp$ ," respectively. The numerical values of the wurtzite GaN lattice parameters are presented in Table I. By comparison of calculations based on Eqs. (2) and (8), we found that the isotropic approximation provides the SPP resonance

frequencies with accuracy of about 1%. This validates the usage of the isotropic model presented in Sec. III.

# C. THz emission from the GaN layer with surface-relief grating

In Subsection IV B, we have shown that the reflectivity spectra of a profiled structure possess the resonance features relating to the excitation of the SPPs. Obviously, one can expect that the emission spectra of the profiled structure will also have the attributes of the SPP excitation. In literature, for a qualitative explanation of the emission properties of the different structures, the theory of the blackbody-like radiation<sup>48</sup> is widely used. In particular, this theory was applied for the analysis of the far-infrared emission of semiconductor films,<sup>49,50</sup> structures with 2D electron gas,<sup>51,52</sup> semiconductor plasmonic structures,<sup>53,54</sup> etc.

The theory of blackbody-like radiation is based on the Kirchhoff's law, which claims that for a body at thermal equilibrium the emissivity is equal to the absorptivity (in particular, for an ideal blackbody, the both quantities are equal to unity). In the case of semiinfinite sample with a planar surface, for a plane incident wave, the mirror reflected radiation is also a plane wave, and the spectral density of blackbody-like radiation emission with given polarization  $\psi$  from the sample which maintained at a temperature *T* can be written as

$$J_{\psi}(\omega,\theta,\phi) = j_{bb}(\omega,T)A_{\psi}(\omega,\theta,\phi)\cos\theta.$$
 (9)

Here,  $j_{bb} = \hbar \omega^3 / 4\pi^3 c^2 [\exp(\hbar \omega / k_B T) - 1]^{-1}$  is the spectral density of radiation emission from an ideal blackbody. The absorptivity of the opaque sample for polarization  $\psi$  is determined as  $A_{\psi}(\omega, \theta, \phi) = 1 - R_{\psi}(\omega, \theta, \phi)$ , where  $R_{\psi}(\omega, \theta, \phi)$  is the reflectivity of the structure,  $\theta$  and  $\phi$  are the polar and azimuth angles, respectively, which specify the propagation direction of the incident wave.

Rigorously, the Kirchhoff's law in the form of Eq. (9) cannot be applied for the considered sample with profiled surface, because generally the surface grating splits the incident wave into several reflected beams. However, in the range of low enough frequencies, when the only diffraction of the zero order takes place, Eq. (9) can be used to estimate the spectral density of the radiation. For that, we define the frequency dependent photoresponse of the detector as follows:

$$U(\omega) = SD(\omega)\Delta\omega[j_{bb}(\omega, T) - j_{bb}(\omega, T_0)]\langle A(\omega)\rangle_{\theta}, \quad (10)$$

where  $T_0$  and T are the epilayer temperature before and just after the voltage pulse,  $D(\omega)$  is spectral sensitivity of the detector, S is the area of the emitting surface, and  $\Delta \omega$  is spectral resolution

$$\langle A(\omega) \rangle_{\theta} = 1/2 \times \int_{\theta_{min}}^{\theta_{max}} \cos \theta [A_p(\omega, \theta, 0) + A_s(\omega, \theta, 0)] d\theta,$$
 (11)

is the absorptivity for unpolarized radiation averaged over incidence angle  $\theta$ . For qualitative description of the experimental emission spectrum, we used here the simplified model with planar geometry. In this model, all the rays lie in the same incident plane which is perpendicular to the grating grooves ( $\phi = 0$ ). Then, averaging the absorptivity for unpolarised radiation over all the polarization angles  $\psi$  gives

$$A(\omega) = 1/(2\pi) \times \int_0^{2\pi} A_{\psi}(\omega, \theta, 0) d\psi$$
  
= 1/2 × [A<sub>p</sub>(\omega, \theta, 0) + A<sub>s</sub>(\omega, \theta, 0)]. (12)

More explicit quantitative description of  $U(\omega)$  which takes into account the real shape of the detected beam can be provided by means of the conical diffraction theory.<sup>55</sup>

To clarify the distinctive THz emission properties related to SPPs, we measured sequentially the spectral dependencies of the detector photoresponse for both the profiled and the reference sample, denoted as  $U(\omega)$  and  $U'(\omega)$ , respectively. The same applied electric power per unit of the sample area was utilized in both cases, which provides approximately the same values of temperature *T* for the both samples. In the framework of the considered model, the ratio  $U(\omega)/U'(\omega)$  is expected to be equal to the ratio  $\langle A(\omega) \rangle_{\theta}/$  $\langle A'(\omega) \rangle_{\theta}$ , where *A'* is the absorptivity of the reference sample. It should be emphasized that the applicability of Eq. (10) which predicts the coincidence of these two ratios is limited in the low frequency range where only diffraction of the zero order takes place.

The experimental results on THz emission from the grating and the reference sample are presented in Fig. 7 by thick line which shows the ratio  $U(\omega)/U'(\omega)$ . Experimental measurements were carried out at  $T_0 = 9$  K under applied electric power of 1500 W. Electrical excitation at this level results in the essential Joule heating of the epitaxial layer. According to our estimation, the effective electron temperature is equal to the epilayer lattice temperature T and reaches a value of ~100 K.

In our experiment, the THz radiation was collected perpendicular to the sample surface in the solid angle  $\Omega = 0.043$  sr (angular aperture  $\Delta \theta = 16^{\circ}$ ). Three peaks of



FIG. 7. THz radiation emission under epilayer temperature modulation. The thick line shows the detector photoresponse for grating sample normalized to photoresponse for reference sample. The thin line presents the ratio  $\langle A(\omega) \rangle_{\theta} / \langle A'(\omega) \rangle_{\theta}$  calculated in the approximation of planar diffraction. The shaded rectangles denote the spectral intervals corresponding to the phase matching condition for SPPs and emitted THz photons at  $\theta = 0^{\circ}$  and  $\Delta \theta = 16^{\circ}$  and different values of factor *M* (see Eq. (3)). The sense of the vertical dashed-dotted lines is given in the caption to Fig. 8.

THz emission with spectral intervals corresponding to the phase matching condition for SPPs and THz photons for  $M = \pm 1, \pm 2$ , and  $\pm 3$  at  $|\theta| \le 8^{\circ}$  were detected. These spectral intervals are marked by shaded rectangles in Fig. 7. Therefore, one can conclude that the characteristic emission peaks originate from the radiative decay of SPP excitations.

The results of the theoretical simulation of the ratio  $\langle A(\omega) \rangle_{\theta} / \langle A'(\omega) \rangle_{\theta}$  also prove this conclusion (see thin line in Fig. 7). The ratio was calculated using Eq. (11) with  $\theta_{min} = -8^{\circ}$  and  $\theta_{max} = 8^{\circ}$  that corresponds to the experimental angular aperture. The calculated curve describes qualitatively the major features of the experimental spectrum. In particular, the calculated positions and widths of the SPP peaks for  $M = \pm 1$  and  $\pm 3$  are very close to the experimentally measured. Moreover, both the experimental and calculated spectra show well-pronounced asymmetric splitting of the emission peaks at frequencies ~3.4 THz and ~10 THz. The origin of this splitting is connected with the finite angle of the detection aperture.

This fact is illustrated in Fig. 8(a) which shows the theoretical spectra of the profiled structure absorptivity calculated at  $\theta = 0^{\circ}$  and  $\theta = 8^{\circ}$  without angular averaging. At  $\theta = 8^{\circ}$ , the splitting of the absorptivity peaks is clearly seen: the number of peaks is twice as large as at  $\theta = 0^{\circ}$ . There are



FIG. 8. Panel (a): Absorptivity spectra for the sample with profiled surface calculated without angular averaging for normal incidence and oblique incidence. Inset: Enlarged image of the spectra in the region of second-order SPP resonance. Panel (b): Angle-averaged absorptivity spectra  $\langle A \rangle_{\theta}$  calculated for  $\theta_{min} = -8^{\circ}$  and  $\theta_{max} = 8^{\circ}$  for the sample with profiled surface (solid line) and reference sample with planar surface (dashed line). Characteristic frequencies of the absorptivity peaks corresponding to SPP resonances of the orders  $M = \pm 1, \pm 2, \pm 3$  at  $\theta = 0^{\circ}$  are shown by the vertical dashed-dotted lines.

four major peaks, namely, sharp absorption peaks at frequencies 3 THz and 4 THz corresponding to M = -1, +1, and broadened peaks at 9 THz and 11.3 THz corresponding to M = -3, +3. After averaging over the angle  $\theta$  (see Fig. 8(b)), the sharp features of the absorption spectrum are smoothed but remain visible (see the solid line in Fig. 8(b)), resulting in asymmetric splitting of the emission peaks in the calculated curve for the ratio  $\langle A(\omega) \rangle_{\theta} / \langle A'(\omega) \rangle_{\theta}$  shown in Fig. 7. However, the theoretical calculations of the absorptivity ratio  $\langle A(\omega) \rangle_{\theta} / \langle A'(\omega) \rangle_{\theta}$  do not reproduce sufficiently strong experimental emission peak near the frequency of the second-order SPPs ( $M = \pm 2$ ). Our theoretical model predicts the strong suppression of the second-order SPP resonance for the symmetric grating with ratio of the  $w_g:a_g = 1:2$  under the normal incidence of radiation.

Similar result was also obtained in the study of the 2D plasmon excitation in semiconductor structures with subwavelength metallic grating.<sup>53,56</sup> Apparently, this mismatch is due to the fact that proposed theory of the planar diffraction incorrectly describes the interaction of the profiled structure with *EM* waves that have non-zero azimuth angle of the incidence plane. Also, the mismatch can be connected with the essential restrictions of the applicability of the Kirchhoff's law for the sample with profiled surface.

#### V. SUMMARY

The theoretical and experimental studies of the interaction of THz radiation with heavily doped GaN epitaxial layers with planar and profiled surfaces are presented. Spectra of the reflectivity and emissivity in wide frequency range are measured and simulated. The measured reflectivity spectra of the sample with planar surface are almost the same for *s*- and *p*-polarization of radiation and show a strong dispersion at the frequencies corresponding to the lowfrequency plasmon-phonon mode (at  $\omega \simeq 0.9 \times \omega_{TO}$ ). The simulations of the reflectivity spectra were carried out using standard formulas for plane-parallel plate with the scalar complex dielectric permittivity, which includes the contributions of the electron and optical phonon subsystems.

The measured reflectivity spectra of the sample with surface-relief grating drastically differ from the spectra of the sample with planar surface. For the *p*-polarized incident beam, the spectrum of the reflectivity has characteristic dips associated with the *excitation* of the surface plasmon polaritons. The positions, amplitudes, and widths of the resonance dips in the reflectivity spectra are well-described by the frequency dependencies of the zero-order diffraction calculated with modified RCWA technique for Maxwell's equations. The mathematical formalism of the RCWA method and its applicability to the geometry of the experiment were discussed in details.

The terahertz radiation emission from the sample with surface-relief grating has features associated with the *decay* of the SPP excitations of different orders. The formalism of the blackbody-like radiation was applied for the explanation of the measured emissivity spectrum. The calculated angle-averaged absorptivities well describe the experimental results on THz emission in the frequency range up to 5 THz ( $\hbar\omega \leq 20$  meV).

The phenomenological theory based on the calculations of the absorptivity spectra could not explain the emergence of the significant emissivity peak corresponding to second-order SPP resonance at ~7.2 THz ( $\hbar\omega \approx 30 \text{ meV}$ ) in the experiment. We assume that it is probably connected with the essential restrictions of the applicability of (i) theory of the planar diffraction to the interaction of the profiled structure with *EM* waves that have non-zero azimuth angle of incidence plane, and (ii) the Kirchhoff's law for the sample with profiled surface.

The experimental and theoretical results presented in this article provide insights for the development of GaNbased devices aimed to absorb/emit terahertz radiation selectively. The effects of essential light absorption in a narrow spectral and angular intervals analyzed above can find applications in chemical and bio-chemical surface plasmon grating detectors working in THz frequency range. The position of the Wood's anomaly depends on the propagation constant of the plasmon surface wave that is quite sensitive to the variation of the refractive index of the surrounding dielectric. Determination of the position of the anomaly brings information about surrounding medium composition, i.e., serves as a THz optical detector. Additionally, a considerable enhancement of the EM energy in the near-field zone of the grating at the SPP resonance can provide an increasing interaction of bio-objects with radiation. We suggest that this will improve the sensitivity of the devices.

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## APPENDIX: APPLICATION OF MODIFIED RCWA METHOD

In the given reference system (see Fig. 2), the *EM* wave with *p*-polarization has two nonzero components of the electric field,  $\vec{E} \{E_x, 0, E_z\}$  and one component of the magnetic field  $\vec{H} \{0, H_y, 0\}$ . Each actual components of *EM* field are the functions of the both *x* and *z* variables and have to satisfy the Maxwell equations written as follows:

$$\frac{\partial H_y}{\partial z} = ik_0 \varepsilon^*(x, z) E_x;$$
$$\frac{\partial H_y}{\partial x} = -ik_0 \varepsilon^*(x, z) E_z;$$
$$\frac{\partial E_x}{\partial z} - \frac{\partial E_z}{\partial x} = ik_0 H_y.$$
(A1)

According the Floquet theorem, we can search for the solutions in the form of Fourier expansion

$$[H_y(x,z), E_x(x,z)]$$
  
=  $\sum_{m=-\infty}^{\infty} [H_{y,m}(z), E_{x,m}(z)] \exp(i\beta_m x),$  (A2)

where

$$\beta_m = k_0 \sqrt{\varepsilon_d} \sin \theta + 2\pi m / a_g. \tag{A3}$$

Then, system (A1) in the Fourier representation is transformed into the system of the  $4N_x + 2$  ordinary differential equations of the first order

$$\frac{\partial H_{y,m}}{\partial z} = ik_0 \sum_{m'=-N_x}^{N_x} \left[ \varepsilon^*(z) \right]_{m,m'} E_{x,m'},$$

$$\frac{\partial E_{x,m}}{\partial z} = -\frac{i\beta_m}{k_0} \sum_{m'=-N_x}^{N_x} \beta_{m'} \left[ \frac{1}{\varepsilon^*(z)} \right]_{m,m'} H_{y,m'} + ik_0 H_{y,m}.$$
 (A4)

Here the truncation rank  $N_x$  is selected in such way to ensure the convergence of the solution with a given accuracy, elements  $[\varepsilon^*(z)^{\pm 1}]_{m,m'}$  compose the Toeplitz matrixes that are given by the following expressions  $[\varepsilon^*(z)^{\pm 1}]_{m,m'} = \int_0^1 \varepsilon^*(\bar{x}, z)^{\pm 1} \exp[-2\pi i \bar{x} (m-m')] d\bar{x}$  with  $\bar{x} = x/a_g$ . In addition to (A4) the Fourier coefficients of z-component of the electric field is expressed as follows:

$$E_{z,m} = -1/k_0 \times \sum_{m'} \beta_{m'} \left[ \frac{1}{\varepsilon^*(z)} \right]_{m,m'} H_{y,m'}.$$
 (A5)

In the spatially uniform regions I and III,  $[\varepsilon^*(z)^{\pm 1}]_{m,m'}$  $\rightarrow [\varepsilon_{d,c}^{\pm 1}]\delta_{m,m'}$  ( $\delta_{m,m'}$  is the Kronecker symbol), as the result, system (A4) is decomposed into the  $2N_x + 1$  independent second-order differential equations

$$\frac{d^2 H_{y,m}^{l,III}}{dz^2} - \left(\lambda_m^{I,III}\right)^2 H_{y,m}^{l,III} = 0.$$
(A6)

Solution of the latter may be written as follows:

$$H_{y,m}^{I} = \delta_{m,0} \exp(-\lambda_{m}^{I} z) + H_{y,m}^{R} \exp(\lambda_{m}^{I} z),$$
  

$$H_{y,m}^{III} = H_{y,m}^{T} \exp(-\lambda_{m}^{III} (z - h_{g})),$$
(A7)

where the incident wave is taken to have amplitude 1,  $\lambda_m^{I,III} = -i\sqrt{\varepsilon_{d,c}k_0^2 - \beta_m^2}$  if  $\operatorname{Re}[\beta_m - \sqrt{\varepsilon_{d,c}}k_0] < 0$  and  $\lambda_m^{I,III}$   $= \sqrt{\beta_m^2 - \varepsilon_{d,c}k_0^2}$  if  $\operatorname{Re}[\beta_m - \sqrt{\varepsilon_{d,c}}k_0] > 0$ . In the first case, when parameter  $\lambda_m$  is the pure imaginary for the several *m*,  $H_{y,m}^{R,T}$  give the amplitudes of reflected and transmitted nonlocalized waves corresponding to *m*-th diffraction orders. In the second case, for all *m* at which  $\operatorname{Re}[\lambda_m^{I,III}] > 0$ ,  $H_{y,m}^{R,T}$ give the amplitudes of reflected and transmitted waves which localized at the surface (evanescent waves). The corresponding components of the electric field are given by

$$E_{x,m}^{I} = \frac{\delta_{m,0} \cos \theta}{\sqrt{\varepsilon_{d}}} \exp\left(-\lambda_{m}^{I}z\right) + \frac{\lambda_{m}^{I}}{ik_{0}\varepsilon_{d}}H_{y,m}^{R}\exp\left(\lambda_{m}^{I}z\right),$$

$$E_{z,m}^{I} = -\frac{\delta_{m,0} \sin \theta}{\sqrt{\varepsilon_{d}}}\exp\left(-\lambda_{m}^{I}z\right) - \frac{\beta_{m}}{k_{0}\varepsilon_{d}}H_{y,m}^{R}\exp\left(\lambda_{m}^{I}z\right),$$

$$E_{x,m}^{III} = -\frac{\lambda_{m}^{III}}{ik_{0}\varepsilon_{c}}H_{y,m}^{T}\exp\left(-\lambda_{m}^{III}(z-h_{g})\right),$$

$$E_{z,m}^{III} = -\frac{\beta_{m}}{k_{0}\varepsilon_{c}}H_{y,m}^{T}\exp\left(-\lambda_{m}^{III}(z-h_{g})\right).$$
(A8)

In the nonuniform region II instead of the conventional solution of the system of the coupled-wave equations (A4) by calculating the eigenvalues and the eigenvectors we proposed to use formal solutions in the frames of the Runge-Kutta methods. In general, such modification of the RCWA technique allows to consider the surface grating with non-uniform distribution of the dielectric permittivity along z-direction. For this purpose it is convenient to rewrite system (A4) in the compact matrix form

$$\frac{d\vec{\mathbf{H}}}{dz} = \hat{\mathbf{A}}(z)\vec{\mathbf{E}},$$

$$\frac{d\vec{\mathbf{E}}}{dz} = \hat{\mathbf{B}}(z)\vec{\mathbf{H}},$$
(A9)

where  $\vec{\mathbf{H}}\{H_{y,m}(z)\}\$  and  $\vec{\mathbf{E}}\{E_{x,m}(z)\}\$  are the vectors of the dimension  $2N_x + 1$ . Matrixes  $\hat{\mathbf{A}}$  and  $\hat{\mathbf{B}}$  have the dimensions  $(2N_x + 1)$  $\times (2N_x + 1)\$  and contain the following elements,  $\{ik_0[\varepsilon^*(z)^{-1}]_{m,m'}^{-1}\}\$  and  $\{ik_0(\delta_{m,m'} - \beta_m\beta_{m'}[\varepsilon^*(z)]_{m,m'}^{-1}/k_0^2)\}\$ , respectively. Here, we used the substitutions  $[\varepsilon^*(z)]_{m,m'} \rightarrow [\varepsilon^*(z)^{-1}]_{m,m'}^{-1}\$  and  $[\varepsilon^*(z)^{-1}]_{m,m'}^{-1} \rightarrow [\varepsilon^*(z)]_{m,m'}^{-1}$  which are proposed and discussed by Li in Ref. 32. In Ref. 32, it was shown that the application of such substitutions is mathematically correct and drastically improves the convergence of the numerical procedure.

Following the conventional Runge-Kutta method of the fourth order (RK4), the solutions of system (A9) in the point  $z_{j+1}$  and  $z_j$  are related to each other as follows:

$$\begin{pmatrix} \vec{\mathbf{H}}(z_{j+1}) \\ \vec{\mathbf{E}}(z_{j+1}) \end{pmatrix} = \hat{\mathbf{S}}(z_j) \begin{pmatrix} \vec{\mathbf{H}}(z_j) \\ \vec{\mathbf{E}}(z_j) \end{pmatrix},$$
(A10)

where

$$\hat{\mathbf{S}} = \begin{pmatrix} \hat{\mathbf{I}} + \frac{1}{2!} \hat{\mathbf{A}} \hat{\mathbf{B}} \Delta z^2 + \frac{1}{4!} \hat{\mathbf{A}} \hat{\mathbf{B}} \hat{\mathbf{A}} \hat{\mathbf{B}} \Delta z^4, & \hat{\mathbf{A}} \Delta z + \frac{1}{3!} \hat{\mathbf{A}} \hat{\mathbf{B}} \hat{\mathbf{A}} \Delta z^3 \\ \hat{\mathbf{B}} \Delta z + \frac{1}{3!} \hat{\mathbf{B}} \hat{\mathbf{A}} \hat{\mathbf{B}} \Delta z^3, & \hat{\mathbf{I}} + \frac{1}{2!} \hat{\mathbf{B}} \hat{\mathbf{A}} \Delta z^2 + \frac{1}{4!} \hat{\mathbf{B}} \hat{\mathbf{A}} \hat{\mathbf{B}} \hat{\mathbf{A}} \Delta z^4 \end{pmatrix}.$$
(A11)

 $\hat{\mathbf{I}}$  is the identity matrix, step size  $\Delta z$  is equal to  $h_g/N_z$  (it is assumed the equidistant partition). Starting from the point  $z_0 = 0$  and applying sequentially  $N_z$  times operator (matrix)  $\hat{\mathbf{S}}(z_j)$  to both left and right parts of Eq. (A10), it is easy to obtain the relationship between vectors of the Fourier coefficients of the transmitted,  $\vec{\mathbf{H}}^T \{ H_{y,m}^T \}$ , and reflected,  $\vec{\mathbf{H}}^R \{ H_{y,m}^R \}$ , waves

$$\begin{pmatrix} \vec{\mathbf{H}}^T \\ \hat{\boldsymbol{\Lambda}}^T \vec{\mathbf{H}}^T \end{pmatrix} = \begin{pmatrix} \hat{\mathbf{T}}_{11}, & \hat{\mathbf{T}}_{12} \\ \hat{\mathbf{T}}_{21}, & \hat{\mathbf{T}}_{22} \end{pmatrix} \begin{pmatrix} \vec{\mathbf{H}}^R + \vec{\delta}_{m,0} \\ \hat{\boldsymbol{\Lambda}}^R \vec{\mathbf{H}}^R + \frac{\cos(\theta_i)}{\sqrt{\varepsilon_1}} \vec{\delta}_{m,0} \end{pmatrix},$$
(A12)

where  $\hat{\mathbf{T}}_{11}$ ,  $\hat{\mathbf{T}}_{12}$ ,  $\hat{\mathbf{T}}_{21}$ ,  $\hat{\mathbf{T}}_{22}$  blocks of the matrix  $\hat{\mathbf{T}} = \prod_{j=1}^{N_z} \hat{\mathbf{S}}(z_j)$  and diagonal matrixes  $\hat{\boldsymbol{\Lambda}}^T$  and  $\hat{\boldsymbol{\Lambda}}^R$  are formed by the elements  $\{\lambda_m^I \delta_{m,m'}/ik_0 \varepsilon_d\}$  and  $\{-\lambda_m^{III} \delta_{m,m'}/ik_0 \varepsilon_c\}$ , respectively.

After some algebraic transformation system (A12) can be rewritten in the form convenient to numerical calculations

$$\begin{pmatrix} \hat{\mathbf{I}}, & -\left(\hat{\mathbf{T}}_{11} + \hat{\mathbf{T}}_{12}\hat{\Lambda}^{R}\right) \\ \hat{\Lambda}^{T}, & -\left(\hat{\mathbf{T}}_{21} + \hat{\mathbf{T}}_{22}\hat{\Lambda}^{R}\right) \end{pmatrix} \begin{pmatrix} \vec{\mathbf{H}}_{R}^{T} \\ \vec{\mathbf{H}}^{R} \end{pmatrix} = \begin{pmatrix} \left(\hat{\mathbf{T}}_{11} + \hat{\mathbf{T}}_{12}\frac{\cos\theta}{\sqrt{\varepsilon_{d}}}\right)\vec{\delta}_{m,0} \\ \left(\hat{\mathbf{T}}_{21} + \hat{\mathbf{T}}_{22}\frac{\cos\theta}{\sqrt{\varepsilon_{d}}}\right)\vec{\delta}_{m,0} \end{pmatrix}.$$
 (A13)

In the case of *s*-polarization, in general, the calculation method remains the same except of some modification of the matrix elements. Now, the non-zero components of *EM* wave are  $H_x$ ,  $H_z$  and  $E_y$ . In system (A9), vectors  $\vec{\mathbf{H}}$  and  $\vec{\mathbf{E}}$  contain elements  $\{H_{x,m}(z)\}$  and  $\{E_{y,m}(z)\}$ , respectively, and matrixes  $\hat{\mathbf{A}}$  and  $\hat{\mathbf{B}}$  are composed by the elements  $\{ik_0(\beta_m^2 \delta_{m,m'}/k_0^2 - [\varepsilon^*(z)]_{m,m'})\}$  and  $\{-ik_0 \delta_{m,m'}\}$ , respectively. Final system of the algebraic equations (A13) should be rewritten as follows:

$$\begin{pmatrix} \hat{\Lambda}^{T}, & -\hat{\mathbf{T}}_{11}\hat{\Lambda}^{R} + \hat{\mathbf{T}}_{12} \\ \hat{\mathbf{I}}, & -\hat{\mathbf{T}}_{21}\hat{\Lambda}^{R} + \hat{\mathbf{T}}_{22} \end{pmatrix} \begin{pmatrix} \vec{\mathbf{E}}^{T} \\ \vec{\mathbf{E}}^{R} \end{pmatrix}$$
$$= \begin{pmatrix} \left( \hat{\mathbf{T}}_{12} - \hat{\mathbf{T}}_{11} \frac{\cos\theta}{\sqrt{\varepsilon_{d}}} \right) \vec{\delta}_{m,0} \\ \left( \hat{\mathbf{T}}_{22} - \hat{\mathbf{T}}_{21} \frac{\cos\theta}{\sqrt{\varepsilon_{d}}} \right) \vec{\delta}_{m,0} \end{pmatrix}, \qquad (A14)$$

with diagonal matrixes  $\hat{\Lambda}^T \{\lambda_m^I \delta_{m,m'}/ik_0 \varepsilon_d\}$  and  $\hat{\Lambda}^R \{\lambda_m^{III} \delta_{m,m'}/ik_0 \varepsilon_c\}$ . Vectors  $\vec{\mathbf{E}}^T$  and  $\vec{\mathbf{E}}^R$  contain the Fourier coefficients of the  $E_y$  component of the transmitted (at  $z=h_g$ ) and reflected (at z=0) waves.

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