Strain-induced three-photon effects

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Strain-induced three-photon effects such as optical second-harmonic generation and hyper-Rayleigh light scattering, characterized by electromagnetic radiation at the double frequency of an incident light, are phenomenologically investigated by adopting a nonlinear photoelastic interaction. The relations between the strain and the nonlinear optical susceptibility for crystal surfaces with point symmetries of $4mm$ and $3m$ are described by a symmetry analysis of the nonlinear photoelastic tensor. We theoretically demonstrate a possibility of determining the strain components by measuring the rotational anisotropy of radiation at the second-harmonic frequency. Hyper-Rayleigh light scattering by dislocation strain is also described using a nonlinear photoelastic tensor. The angular dependencies of light scattered at the double frequency of an incident light for different scattering geometries are analyzed.

I. INTRODUCTION

The methods of photoelasticity in linear optics are well developed for the investigation of strain in solids.[1,2] External strain applied to a solid leads to a change in the shape and symmetry of a specimen.[3] Internal strain in composite materials—for example, in crystal films on substrates or multilayers—leads to a lowering of the symmetry near the interfaces and to structural deformations induced by a misfit between crystal lattice constants.[4] The influence of strain on the optical properties of solids in the phenomenological framework can be described by introducing a photoelastic term in the dielectric permittivity tensor as[1,2]

$$\epsilon_{ij} = \epsilon_{ij}^{(0)} + p_{ijkl}u_{kl}, \qquad (1)$$

where $\epsilon_{ij}^{(0)}$ is the dielectric permittivity tensor of a nondeformed medium, and p_{ijkl} and u_{kl} are the linear photoelastic tensor and the strain tensor, respectively.

Nonlinear optical methods are widely used for the investigation of real solids, and are usually more powerful than linear optical methods.[5] Therefore, it is necessary to extend the possibilities of photoelasticity for the case of three-wave optical interactions. Optical second-harmonic generation (SHG) is very efficient for an investigation of thin films and interfaces.[5–7] The SHG signal is very sensitive to a change of symmetry induced by various physical actions—for example, by strain. Special kinds of internal strain (not hydrostatic) lead to a lowering of the symmetry of a crystal, i.e., to a missing of the inversion center.[3] As a result, bulk dipole-active optical SHG should be observed for transparent materials in the transmission geometry. From another side, surfaces and interfaces of centrosymmetric bulk materials are characterized by a lower point-symmetry group even in the absence of strain. In this case a more effective method to observe strain-induced changes of a surface is the measurement of optical SHG in the reflection geometry. In both cases, a method using three-photon phenomena is an effective tool for investigating strain-induced effects in solids and solid-state structures. A number of publications[8–19] were devoted to both theoretical and experimental investigations of a strain influence on SHG in Si films[8–12,17,19] and GaN films[15,18] on different substrates (see also references in the recent review article[20]). However, these studies were concerned only with calculations and measurements of strain-induced resonances in the second harmonic spectra.

Real crystals contain defects which are the sources of the long-range strain fields around the structural inhomogeneities, for example, such as a dislocations.[4,21] These defect-induced strains also change the nonlinear optical susceptibility (NOS) of a crystal, and lead to nonlinear elastic scattering or hyper-Rayleigh light scattering (HRLS).[22] Both three-photon effects, SHG and HRLS, are characterized by radiation at the double frequency of an incident light. However, radiation corresponding to the HRLS, propagates in an arbitrary direction, whereas for the SHG it is necessary to satisfy the phase-matching conditions,[5] or specular reflection (in the case of surfaces or nontransparent films). HRLS has been applied for the investigation of inhomogeneous crystals of KH_2PO_4,[23] $Sr_{1-x}Ca_xTaO_3$,[24] and $K_{1-x}Li_xTaO_3$,[25] as well as of inhomogeneous solid films of bacteriorodopsin,[26] polycrystalline ferroelectric films of $Pb_x(Zr_{0.53}Ti_{0.47})O_3$,[26] and Langmuir film of C_{60}.[27] Recently, a phenomenological model describing the influence of dislocations on the second-order NOS tensor was developed by Bottomley.[28] However, in our opinion, this approach does not give an adequate description of the influence of the dislocation strain on the nonlinear elastic light scattering even with the correction made in Ref. 29. A more realistic description of the influence of dislocation strain on three-photon effects involves the introduction of a nonlinear photoelastic interaction simililar to

the model proposed by Nelson and Lax for the description of the acoustically induced optical SHG.[30]

In this paper, we present a phenomenological description of strain-induced optical SHG and group-theoretical analysis of the second-order NOS tensor, taking into account a nonlinear photoelastic interaction like the approach developed in Ref. 30. Generally speaking, for both ''reflection'' and ''transmission'' geometries of SHG observation, the bulk and surface second-order nonlinear optical polarizations are the sources of radiation at the double frequency of an incident light. As pointed by Bottomley et al.,[31] it is possible to separate the bulk and surface contributions to the SHG via measurement of rotational anisotropy at the second-harmonic frequency, because these sources of nonlinear polarizations are characterized by different point-symmetry groups. We consider the influence of strain on the s-polarized surface optical SHG for the $4mm$ and $3m$ surfaces of a cubic crystal, neglecting the bulk contribution to the effect. Nevertheless, general formulas in this paper are also applicable to describe bulk strain-induced optical SHG. As an example, we investigate HRLS caused by strain induced by the straight edge dislocation.

This paper is organized as follows. In Sec. II we consider general relationships for describing strain-induced changes of nonlinear polarization and the second-order NOS tensor, respectively. In this section we also analyze the change of the surface symmetry under different kinds of strain, and discuss the possibility of an observation of this changes via measurements of rotational anisotropy of the SHG. In Sec. III, we investigate HRLS by the dislocation strain in the framework of the general approach of nonlinear photoelasticity developed in Sec. II. Finally, in Sec. IV we summarize the results obtained in the present paper and outline future perspectives of strain-induced nonlinear optics in ordered media.

II. NONLINEAR PHOTOELASTIC TENSOR

The second-order nonlinear optical polarization $\mathbf{P}^{\mathrm{ND}(2)}(2\omega)$ at the double frequency of the incident light in the dipole approximation can be written in the well-known form[5]

$$P_i^{\mathrm{NL}}(2\omega) = \chi_{ijk}^{(2)}(-2\omega:\omega,\omega)E_j(\omega)E_k(\omega), \quad (2)$$

where $\chi_{ijk}^{(2)}$ is the second-order NOS tensor, and $\mathbf{E}(\omega)$ is the electric field of the incident light at the frequency ω. Within the phenomenological approach an influence of strain on the second-order nonlinear polarization can be described by the nonlinear photoelastic tensor. The nonlinear photoelastic interaction was used in the description of acoustically induced optical SHG (Refs. 30 and 32) and high-order Brillouin light scattering by acoustical phonons.[33] Similar to Eq. (1), the strain effect on the second-order NOS tensor in the linear approximation can be presented as

$$\chi_{ijk}^{(2)} = \chi_{ijk}^{(2,0)} + p_{ijklm}u_{lm}, \quad (3)$$

where $\chi_{ijk}^{(2,0)}$ is the second-order NOS tensor of unstrained crystal, and p_{ijklm} is the nonlinear photoelastic tensor. The authors of Ref. 32 determined the nonzero components of second-order NOS and photoelastic tensors in GaAs in the

infrared region (with wavelengths $\lambda_\omega = 10.6\,\mu\mathrm{m}$ and $\lambda_{2\omega} = 5.3\,\mu\mathrm{m}$) and found that these values are comparable: $\chi_{xyz}^{(2,0)} = 1.3 \times 10^{-10}$ m/V and $|p_{xxxyz}| \simeq 1.2 \times 10^{-10}$ m/V.

From Eq. (3) it follows that the symmetry properties of the nonlinear photoelastic tensor p_{ijklm} are given by the symmetry properties of the NOS tensor and the strain tensor. By definition, the strain tensor u_{lm} is symmetric, i.e., $u_{lm} = u_{ml}$. As follows from Eq. (2), the second-order NOS tensor $\chi_{ijk}^{(2)}$ is symmetric between two indices (j,k), i.e., $\chi_{ijk}^{(2)} = \chi_{ikj}^{(2)}$ (the same is valid for $\chi_{ijk}^{(2,0)}$). Therefore, the nonlinear photoelastic tensor p_{ijklm} is symmetric between two pairs of indices (j,k) and (l,m), i.e., $p_{ijklm} = p_{ikjlm} = p_{ijkml} = p_{ikjml}$. These relationships result in a reduction of the number of independent components of each tensor.

All components of odd rank tensors $\chi_{ijk}^{(2,0)}$ and p_{ijklm} are equal to zero in centrosymmetric crystals. However, surfaces and interfaces of centrosymmetric bulk materials are characterized by lower point-symmetry groups. For example, (001) and (111) surfaces of the cubic crystal are described by $4mm$ and $3m$ point-symmetry groups, respectively. In these cases the nonzero components of odd rank tensor $\chi_{ijk}^{(2,0)}$ and p_{ijklm} can be easily calculated (see Table I).[34]

From Eqs. (2) and (3) the general expressions for the components of nonlinear polarization in a stressed medium can be expressed in terms of the NOS tensor, the strain tensor, and the nonlinear photoelastic tensor. For a simple (001) surface of cubic crystal the nonlinear polarization is determined as follows:

$$P_x^{\mathrm{NL}}(2\omega) = 2[p_{xxxxz}E_x^2(\omega) + p_{xyyxz}E_y^2(\omega) + p_{xzzxz}E_z^2(\omega)]u_{xz}$$
$$+ 4[p_{xyxyz}u_{yz}E_y(\omega)E_x(\omega)$$
$$+ p_{xyzxy}u_{xy}E_y(\omega)E_z(\omega)]$$
$$+ [2\chi_{xxz}^{(2,0)} + 2(p_{xxzxx}u_{xx} + p_{xxzyy}u_{yy}$$
$$+ p_{xxzzz}u_{zz})]E_x(\omega)E_z(\omega), \quad (4)$$

$$P_y^{\mathrm{NL}}(2\omega) = 2[p_{yyyyz}E_y^2(\omega) + p_{yxxyz}E_x^2(\omega)$$
$$+ p_{yzzsyz}E_z^2(\omega)]u_{yz} + 4[p_{yxyxz}u_{xz}E_x(\omega)E_y(\omega)$$
$$+ p_{yxzyx}u_{xy}E_x(\omega)E_z(\omega)]$$
$$+ [2\chi_{yyz}^{(2,0)} + 2(p_{yyzyy}u_{yy} + p_{yyzxx}u_{xx}$$
$$+ p_{yyzzz}u_{zz})]E_y(\omega)E_z(\omega), \quad (5)$$

$$P_z^{\mathrm{NL}}(2\omega) = (\chi_{zxx}^{(2,0)} + p_{zxxxx}u_{xx} + p_{zxxyy}u_{yy} + p_{zxxzz}u_{zz})E_x^2(\omega)$$
$$+ (\chi_{zyy}^{(2,0)} + p_{zyyxx}u_{xx} + p_{zyyyy}u_{yy}$$
$$+ p_{zyyzz}u_{zz})E_y^2(\omega) + (\chi_{zzz}^{(2,0)} + p_{zzzxx}u_{xx}$$
$$+ p_{zzzyy}u_{yy} + p_{zzzzz}u_{zz})E_z^2(\omega)$$
$$+ 4[p_{zxyyx}u_{xy}E_x(\omega)E_y(\omega)$$
$$+ p_{zxzxz}u_{xz}E_x(\omega)E_z(\omega)$$
$$+ p_{zyzyz}u_{yz}E_y(\omega)E_z(\omega)]. \quad (6)$$

From Eqs. (4)–(6) and Table I we can determine the changes in nonlinear polarization under different kinds of strain.

TABLE I. Nonzero components of the nonlinear photoelastic tensor p_{ijklm} [p_{ijklm} is symmetric between two pairs of indices (j,k) and (l,m)].

Crystal symmetry	Nonzero components of p_{ijklm}
$4mm$	$yxxyz=xyyxz$, $yxyxz=xxyyz$, $yxzxy=xyzyx$, $yyyyz=xxxxz$, $yyzxx=xxzyy$
	$yyzyy=xxzxx$, $yyzzz=xxzzz$, $yzzyz=xzzxz$, $zxyyx$, $zyyxx=zxxyy$
	$zyyyy=zxxxx$, $zyyzz=zxxzz$, $zyzyz=zxzxz$, $zzzyy=zzzxx$, $zzzzz$
$3m$	$xxxyy$, $xxxx$, $yyyyx=yxxxy=-\frac{1}{2}xxxxx+\frac{1}{2}xxxyy$
	$xyyy=xyyxx=xxxx$, $yyxyy=yyxxx=-\frac{1}{2}xxxxx-\frac{1}{2}xxxyy$
	$xyxyz=-yyxxz$, $xxxxz=-yyyyz$, $yxxyz=xyyxz=xxxxz-2xyxz$
	$xxxzz=-yyxzz=-xyyzz$, $xyzxz=yxzyx=\frac{1}{2}xxzxz-\frac{1}{2}xxzyy$
	$xxzxx=yyzyy$, $yyzzx=yxzzy=xyzzy=-xxzxz$, $xzzxx=-yzzyx=-xzzyy$
	$zxyxy=\frac{1}{2}zxxxx-\frac{1}{2}zxxyy$, $zxxyy=zyyxx$, $zzzyy=zzzxx$
	$zxxxz=-zyyxz=-zxyyz$, $zxzxx=-zyzyx=-zxzyy$, $yyzzz=xxzzz$
	$xxzyy=yyzxx$, $yzzyz=xzzxz$, $zyyzz=zxxzz$, $zyzyz=zxzxz$, $zzzzz$

Table II summarizes the relations between the components of the second-order NOS tensor $\chi_{ijk}^{(2)}$ and the strain tensor u_{lm} for $4mm$ and $3m$ symmetry classes.

Strain alters the physical properties of a crystal, and leads to a lowering of symmetry. For example, the (001) surface of a cubic crystal has a $4mm$ tetragonal symmetry and changes into a $mm2$ orthohombic symmetry with application of the appropriate stress. Figure 1 illustrates the possible changes of the symmetry due to stress. This symmetry lowering is clearly seen in the rotational anisotropy of the SHG intensities. For an investigation of rotational anisotropy of the SHG, we consider the relationships between the second-order NOS tensor $\chi_{i'j'k'}^{(2)}$ and $\chi_{ijk}^{(2)}$ in the beam coordinates $x'y'z'$ connected with incident and scattered light, and in the crystal coordinates xyz connected with the specimen. When z and z' axes coincide, and x' and y' axes are rotated by the angle ϕ relative to the x and y axes, respectively, the s-polarized component of SHG for both s- and p-polarized incident light can be expressed as follows:

$$P_{s(\omega)\to s(2\omega)}^{NL}=\chi_{y'y'y'}^{(2)}E_{y'}^2(\omega)$$
$$=[-\chi_{xxx}^{(2)}\sin^3\phi+\chi_{yyy}^{(2)}\cos^3\phi$$
$$+(2\chi_{xxy}^{(2)}+\chi_{yxx}^{(2)})\sin^2\phi\cos\phi$$
$$-(2\chi_{yyx}^{(2)}+\chi_{xyy}^{(2)})\cos^2\phi\sin\phi]E_{y'}^2(\omega), \quad (7)$$

$$P_{p(\omega)\to s(2\omega)}^{NL}=\chi_{y'x'x'}^{(2)}E_{x'}^2(\omega)+2\chi_{y'x'z'}^{(2)}E_{x'}(\omega)E_{z'}(\omega)$$
$$+\chi_{y'z'z'}^{(2)}E_{z'}^2(\omega)$$
$$=[(2\chi_{yyy}^{(2)}-\chi_{xxx}^{(2)})\cos^2\phi\sin\phi$$
$$+(\chi_{yyy}^{(2)}-2\chi_{xxy}^{(2)})\sin^2\phi\cos\phi+\chi_{yxx}^{(2)}\cos^3\phi$$
$$-\chi_{xyy}^{(2)}\sin^3\phi]E_{x'}^2(\omega)$$
$$+[(\chi_{yyz}^{(2)}-\chi_{xxz}^{(2)})\sin\phi\cos\phi+\chi_{yxz}^{(2)}\cos^2\phi$$
$$-\chi_{xyz}^{(2)}\sin^2\phi]E_{x'}(\omega)E_{z'}(\omega)$$
$$+[\chi_{yzz}^{(2)}\cos\phi-\chi_{xzz}^{(2)}\sin\phi]E_{z'}^2(\omega). \quad (8)$$

Equations (7) and (8) are the general formulas for any crystal symmetry class. These can be simplified for a specific point-symmetry class of a medium. Substituting the nonzero components of the second-order NOS tensor $\chi_{ijk}^{(2)}$ into Eqs. (7) and (8), we can determine the nonlinear polarization for the $4mm$, $mm2$, $3m$, and m symmetry classes (see Table III). For these various cases the intensities of SHG are illustrated in polar coordinates in Fig. 2. It can be seen from these figures that the SHG intensities definitely reflect the symmetry of a nonlinear medium. Thus symmetry lowering induced by strain can also be found in the rotational anisotropy of the SHG intensity.

TABLE II. The relation between the strain and the NOS tensor components in $4mm$ and $3m$ symmetry classes. The NOS tensor components represented by the bold characters are equal to zero in a strain-free crystal. Because of nonszero NOS tensor components, the crystal no longer has $4mm$ or $3m$ symmetry.

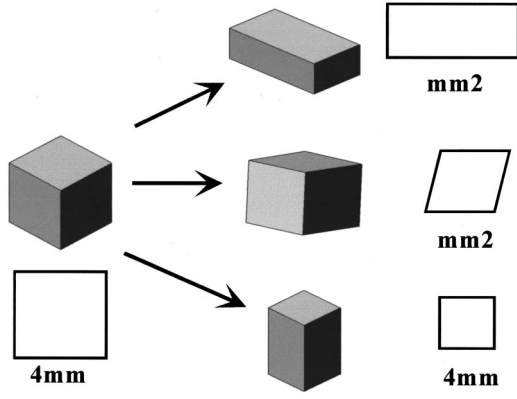| Components of strain tensor, u_{lm}	Components of second-order NOS tensor, $\chi_{ijk}^{(2)}$	
	$4mm$	$3m$
xx,yy,zz	$xxz=xzx$, $yyz=yzy$, zxx, zyy, zzz,	xxx, $xxz=xzx$, xyy, $yxy=yyx$, $yyz=yzy$,
$xz=zx$	**xxx, xyy, xzz, yxy=yyx, zxz=zzx**	zxx, $\mathbf{xzz}$, $\mathbf{zxz=zzx}$, zyy, $\mathbf{zzx=zxz}$, zzz
$yz=zy$	**xxy=xyx, yxx, yyy, yzz, zyz=zzy**	**xxy=xyx, xyz=xzy, yxx, yxz=yzx,**
$xy=yx$	**xyz=xzy, yxz=yzx, zxy=zyx**	**zxy=zyx, zyz=zzy, yyy, yzz**

FIG. 1. The symmetry changes of a $4mm$ crystal due to internal strain.

The symmetry class of a crystal imposes restrictions on the form of the second-order NOS tensor. For the $4mm$ symmetry class the second-order NOS tensor is characterized by the nonzero components[34] $\chi^{(2,0)}_{xxz}=\chi^{(2,0)}_{yyz}=\chi^{(2,0)}_{xzx}=\chi^{(2,0)}_{yzy}$, $\chi^{(2,0)}_{zxx}=\chi^{(2,0)}_{zyy}$, $\chi^{(2,0)}_{zzz}$. The discrepancy between the relationships of the second-order NOS tensor components for $4mm$ and $mm2$ symmetry classes lies in the interchange of x and y indices. For example, for symmetry class $4mm$ $\chi^{(2,0)}_{xxz}=\chi^{(2,0)}_{yyz}$, but for $mm2$ symmetry class $\chi^{(2,0)}_{xxz}\neq\chi^{(2,0)}_{yyz}$. The difference between $\chi^{(2)}_{xxz}$ and $\chi^{(2)}_{yyz}$ components of the NOS tensor in a strained medium can be described by the strain tensor and the nonlinear photoelastic tensor as follows:

$$\Delta\chi(4mm\rightarrow mm2)=\chi^{(2)}_{xxz}-\chi^{(2)}_{yyz}=(p_{xxzlm}-p_{yyzlm})u_{lm}. \quad (9)$$

Taking into account the nonzero components of the nonlinear photoelastic tensor for $4mm$ symmetry class in Table I, from Eq. (9) we obtain the following expression for $\Delta\chi(4mm\rightarrow mm2)$ ($\chi^{(2,0)}$, p_{ijklm}, and u_{lm} have $4mm$ symmetry, while $\chi^{(2)}$ has $mm2$ symmetry):

$$\Delta\chi(4mm\rightarrow mm2)=(p_{xxzxx}-p_{xxzyy})(u_{xx}-u_{yy}). \quad (10)$$

Naturally, when the symmetry of the medium is changed from $4mm$ to $mm2$ due to strain, the change in the SHG intensity should include the changes of the NOS tensor components in Eqs. (7) and (8). There is no SHG in dipole ap-
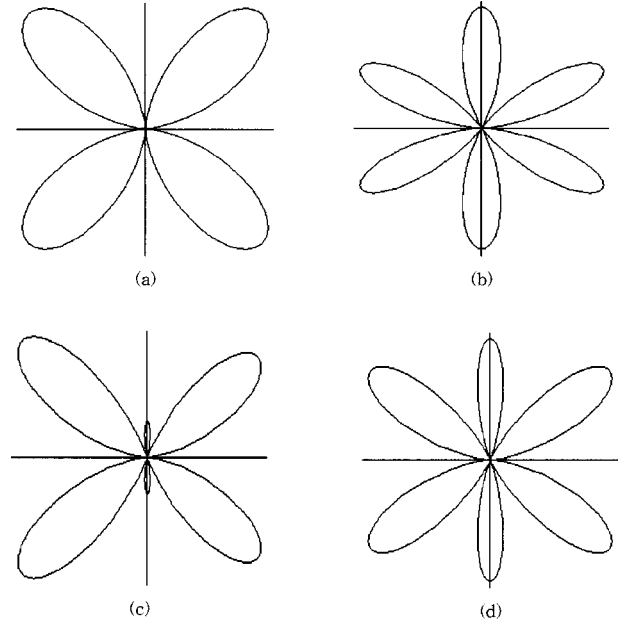

FIG. 2. The rotational anisotropies of the SHG intensities for the $s(\omega)\rightarrow p(2\omega)$ polarization configuration in (a) $mm2$, (b) $3m$, and (c) m crystal surface symmetries, and for the $s(\omega)\rightarrow s(2\omega)$ polarization configuration in (d) the m crystal surface symmetry.

proximations for both $s(\omega)\rightarrow s(2\omega)$ and $p(\omega)\rightarrow s(2\omega)$ configurations for a (001) surface of $4mm$ cubic symmetry. However, when a surface is deformed to $mm2$ symmetry under stress, the SHG intensity $I(2\omega)$ no longer vanishes for $p(\omega)\rightarrow s(2\omega)$ polarization. Taking into account the nonzero components of the second-order NOS tensor and the nonlinear photoelastic tensor from Eqs. (7) and (8), we obtain the following equation:

$$I_{p(\omega)\rightarrow s(2\omega)}(2\omega)\propto|P^{\mathrm{NL}}_{p(\omega)\rightarrow s(2\omega)}(2\omega)|^2$$
$$=\tfrac{1}{4}|\Delta\chi(4mm\rightarrow mm2)|^2$$
$$\times\sin^2 2\phi|E_{x'}(\omega)|^2|E_{z'}(\omega)|^2. \quad (11)$$

From this equation one can clearly see the fourfold rotational anisotropy shown in Fig. 2(a). From Eq. (11) it follows that the amount of the symmetry change in the SHG intensities is proportional to the difference in the nonlinear photoelastic

TABLE III. Nonlinear polarization for $4mm$, $mm2$, $3m$, and m symmetry classes.

Symmetry class	In and out polarizations	Nonlinear polarization, $\mathbf{P}^{\mathrm{NI}}(2\omega)$
$4mm$	$s(\omega)\rightarrow s(2\omega)$	0
	$p(\omega)\rightarrow s(2\omega)$	0
$mm2$	$s(\omega)\rightarrow s(2\omega)$	0
	$p(\omega)\rightarrow s(2\omega)$	$-\tfrac{1}{2}(\chi_{xxz}-\chi_{yyz})E_{x'}(\omega)E_{z'}(\omega)\sin 2\phi$
$3m$	$s(\omega)\rightarrow s(2\omega)$	$\chi_{xxx}E^2_{y'}(\omega)\sin 3\phi$
	$p(\omega)\rightarrow s(2\omega)$	$-\chi_{xxx}E^2_{z'}(\omega)\sin 3\phi$
m	$s(\omega)\rightarrow s(2\omega)$	$-[\chi_{xxx}\sin^2\phi+(\chi_{xyy}+2\chi_{yyx})\cos^2\phi]E^2_{y'}(\omega)\sin\phi$
	$p(\omega)\rightarrow s(2\omega)$	$-[\chi_{xyy}\sin^2\phi+(\chi_{xxx}-2\chi_{yyx})\cos^2\phi]E^2_{x'}(\omega)\sin\phi$
		$-(\chi_{xxz}-\chi_{yyz})E_{x'}(\omega)E_{z'}(\omega)\sin\phi\cos\phi$
		$-\chi_{xzz}\sin\phi E^2_{z'}(\omega)$

TABLE IV. The Fourier coefficients in Eq. (14) for the m symmetry crystal class. All Δ_i's (c_1, c_2, c_4, c_5 also) are equal to zero for $3m$ symmetry. (The asterisk denotes the complex conjugate.)

Fourier coefficient	$s(\omega) \rightarrow s(2\omega)$	$p(\omega) \rightarrow s(2\omega)$
c_0	$\frac{1}{2}(|\chi_{xxx}^{(2)}|^2 - \Delta_2 \chi_{xxx}^{(2)})|E_{y'}(\omega)|^4$	$\frac{1}{4}(2|\chi_{xxx}^{(2)}|^2 - \Delta_1 \chi_{xxx}^{(2)} - 2\Delta_2 \chi_{xxx}^{(2)})|E_{x'}(\omega)|^4$
c_1	0	$-\frac{1}{4}\Delta_3 \chi_{xxx}^{(2)}|E_{x'}(\omega)|^2[E_{x'}(\omega)E_{z'}(\omega) + E_{x'}^*(\omega)E_{z'}^*(\omega)]$
c_2	$-\frac{1}{4}(\Delta_1 + 2\Delta_2)\chi_{xxx}^{(2)}|E_{y'}(\omega)|^4$	$\frac{1}{4}[(3\Delta_1 - 2\Delta_2)|E_{x'}(\omega)|^4 + 4\Delta_4|E_{x'}(\omega)|^2|E_{z'}(\omega)|^2]\chi_{xxx}^{(2)}$
c_3	0	0
c_4	$\frac{1}{4}(\Delta_1 + 2\Delta_2)\chi_{xxx}^{(2)}|E_{y'}(\omega)|^4$	$\frac{1}{4}[(2\Delta_2 - 3\Delta_1)|E_{x'}(\omega)|^4 - 4\Delta_4|E_{x'}(\omega)|^2|E_{z'}(\omega)|^2]\chi_{xxx}^{(2)}$
c_5	0	$\frac{1}{4}\Delta_3 \chi_{xxx}^{(2)}|E_{x'}(\omega)|^2[E_{x'}(\omega)E_{z'}(\omega) + E_{x'}^*(\omega)E_{z'}^*(\omega)]$
c_6	$\frac{1}{4}(\Delta_1 + 2\Delta_2 - 2\chi_{xxx}^{(2)})\chi_{xxx}^{(2)}|E_{y'}(\omega)|^4$	$\frac{1}{4}(\Delta_1 \chi_{xxx}^{(2)} + 2\Delta_2 \chi_{xxx}^{(2)} - 2|\chi_{xxx}^{(2)}|^2)|E_{x'}(\omega)|^4$

tensor components and the diagonal components of u_{xx} and u_{yy} of the strain tensor. The values of nonlinear photoelastic tensor components p_{ijklm} can be determined from independent experiments: for instance, Boyd, Nash, and Nelson determined the nonlinear photoelastic tensor components by measurements of acoustically induced SHG.[32] Thus, if we have information about the original symmetry of a medium before stress is applied and the rotational anisotropy of the SHG intensities, the stress components or the relationships between them associated with the symmetry lowering can be determined.

For a (111) surface of $3m$ symmetry, the relationships for the SHG intensity are more complicated. However, they can be simplified by Fourier transformation. The intensity of the SHG can be written using Fourier coefficients in the following form:

$$I(2\omega) = \sum_{n=0}^{6} c_n \cos(n\phi). \qquad (12)$$

Each Fourier component c_n of the SHG intensity represents the rotational symmetry of the crystal. Similar to the $4mm$ symmetry case considered above, we can find a connection between symmetry lowering and the relationships of the second-order NOS tensor components. Initially, the cubic (111) surface has a sixfold symmetry in SHG intensities [Fig. 2(b)]. This rotational symmetry is broken when strain is applied. If strain breaks the sixfold rotational symmetry while maintaining the mirror symmetry, the following components of the second-order NOS tensor contribute to the SHG for $s(\omega) \rightarrow s(2\omega)$ and $p(\omega) \rightarrow s(2\omega)$ polarization configurations: $\chi_{xxx}^{(2)}$, $\chi_{xyy}^{(2)}$, $\chi_{yxy}^{(2)}$, $\chi_{xxz}^{(2)}$, and $\chi_{yyz}^{(2)}$. For the $3m$ symmetry class, from Ref. 34 we obtain

$$\chi_{xzz}^{(2)} = 0, \quad \chi_{xxx}^{(2)} = -\chi_{xyy}^{(2)} = -\chi_{yxy}^{(2)}, \quad \chi_{yyz}^{(2)} = \chi_{xxz}^{(2)}. \qquad (13)$$

These relationships are also approximately correct in the m symmetry class because the changes of components in linear approximation are usually small. The strain tensor components u_{xx}, u_{yy}, u_{zz}, and u_{xz} affect these second-order NOS tensor components, and we can define the discrepancies Δ_i between relationships (13) of the components of the second-order NOS tensor for $3m$ and m symmetry classes, taking into account the nonzero components of p_{ijklm} (Ref. 34):

$$\Delta_1 = \chi_{xxx}^{(2)} + \chi_{xyy}^{(2)} = p_{xxxjk}u_{jk} + p_{xyylm}u_{lm}$$
$$= (p_{xxxxx} - p_{xxxyy})(u_{xx} - u_{yy})$$
$$+ 4(p_{xxxxz} - p_{xxyyz})u_{xz}, \qquad (14)$$

$$\Delta_2 = \chi_{xxx}^{(2)} + \chi_{yxy}^{(2)} = p_{xxxjk}u_{jk} + p_{yxylm}u_{lm}$$
$$= \frac{1}{2}(p_{xxxxx} - p_{xxxyy})(u_{xx} - u_{yy})$$
$$+ 2(p_{xxxxz} + p_{xxyyz})u_{xz}, \qquad (15)$$

$$\Delta_3 = \chi_{yyz}^{(2)} - \chi_{xxz}^{(2)} = p_{yyzjk}u_{jk} - p_{xxzlm}u_{lm}$$
$$= (p_{xxzyy} - p_{xxzxx})(u_{xx} - u_{yy}) - 4p_{xxzxz}u_{xz}, \qquad (16)$$

$$\Delta_4 = \chi_{xzz}^{(2)} = p_{xzzjk}u_{jk} = p_{xzzxx}(u_{xx} - u_{yy}) + 2p_{xzzxz}u_{xz}. \qquad (17)$$

In these notations, SHG intensities can be divided into each Fourier coefficient for each polarization configuration, as presented in Table IV [also see Figs. 2(c) and 2(d)]. In this table, the first-order terms of Δ_i and the real parts of $\chi_{ijk}^{(2)}$ are included for simplicity. From the rotational anisotropy measurements of the SHG intensity it is possible to find the corresponding Fourier components c_n presented in the Table IV for both cases of $s(\omega) \rightarrow s(2\omega)$ and $p(\omega) \rightarrow s(2\omega)$ polarizations. These values c_n are determined by nonzero components of the second-order NOS tensor and Δ_i, the latter depends only on the nonlinear photoelastic tensor and strain tensor. From the values of c_n, Δ_i in Table IV and Eqs. (14)–(17), $u_{xx} - u_{yy}$, u_{xz} can be calculated. These components are obtainable without detailed information about the unstressed susceptibility $\chi_{ijk}^{(2,0)}$ if the symmetry of the unstressed medium is well known. It should be noted that the other off-diagonal stress components can also be determined from different polarization configurations in a similar manner to that described above.

III. HYPER-RAYLEIGH LIGHT SCATTERING BY DISLOCATION STRAIN

On the other hand, there are some special cases in which the analytic forms of strain field are already identified. It is well known that real crystals contain dislocations which are the source of the long-range strain fields.[4,21] These strains also lead to a change of nonlinear optical properties of the crystal, and can be detected by HRLS. In this section we present a phenomenological description of HRLS by dislocation strain using the approach developed in Sec. II.

Let us consider a cubic crystal without an inversion center, for example GaAs (the point symmetry $\bar{4}3m - T_d$), with the edge dislocation oriented along the z axis with Burgers vector $\mathbf{b} = (b,0,0)$. In the crystallographic coordinate basis xyz, the dislocation strain is characterized by the nonzero components of the strain tensor $u_{ik}(\mathbf{r})$,

$$u_{xx}(\mathbf{r}) = -\frac{b}{4\pi(1-\nu)}\frac{y[x^2(3-2\nu)+y^2(1-2\nu)]}{(x^2+y^2)^2},$$
(18)

$$u_{yy}(\mathbf{r}) = \frac{b}{4\pi(1-\nu)}\frac{y[x^2(1+2\nu)+y^2(1-2\nu)]}{(x^2+y^2)^2},$$
(19)

$$u_{xy}(\mathbf{r}) = \frac{b}{4\pi(1-\nu)}\frac{x(x^2-y^2)}{(x^2+y^2)^2},$$
(20)

where ν is Poisson's coefficient.

Let us determine the polarization of light scattered at the second-harmonic frequency in the same rotational geometry discussed in Sec. II. Within the slowly varing amplitude approximation the wave equation for the second-harmonic electric field can be written as[5]

$$2ik_{2\omega,i}\mathbf{\nabla}_i E_i(2\omega,\mathbf{q}) = -\frac{\omega^2}{c^2}\chi_{ijk}^{(2)}(\mathbf{r})E_j(\omega)E_k(\omega)\exp(i\mathbf{q}\cdot\mathbf{r}),$$
(21)

where $\mathbf{q} = 2\mathbf{k}_\omega - \mathbf{k}_{2\omega}$ is the scattering wave vector, while $\mathbf{k}_\omega$ and $\mathbf{k}_{2\omega}$ are the wave vectors of the fundamental and second harmonic light, respectively. Using the infinite plane-wave approximation,[5] from Eq. (21) we obtain

$$E_i(2\omega,\mathbf{q}) = \frac{A}{V}\int_V \chi_{ijk}^{(2)}(\mathbf{r})E_j(\omega)E_k(\omega)\exp(i\mathbf{q}\cdot\mathbf{r})d\mathbf{r},$$
(22)

where $A = -i\omega/cn_{2\omega}$. The integral in Eq. (22) is taken over the interaction volume V, and $n_{2\omega}$ is the refractive index of the crystal at the second-harmonic frequency.

Substituting the nonzero components of the nonlinear photoelastic tensor for the point-symmetry group $43m$ and the dislocation stran tensor determined by Eqs. (18)–(20) into Eq. (22), we obtain the values of the electric fields at the second-harmonic frequency for the s- and p-polarized incident light as follows:

(i) $s(\omega) \rightarrow s(2\omega)$: $E_{y'}(2\omega,\mathbf{q}) = 0$;   (23)

(ii) $p(\omega) \rightarrow s(2\omega)$:

$$E_{y'}(2\omega,\mathbf{q}) = A\Big\{\chi_{yzx}^{(2,0)}f(\mathbf{q})\cos 2\phi + p_{yxzyy}$$
$$\times[u_{yy}(\mathbf{q})\cos^2\phi - u_{xx}(\mathbf{q})\sin^2\phi]$$
$$+\frac{1}{2}(p_{xxzxy}-p_{yyzxy})u_{xy}(q)\sin 2\phi\Big\}$$
$$\times E_{x'}(\omega)E_{z'}(\omega);$$
(24)

(iii) $s(\omega) \rightarrow p(2\omega)$: $E_{x'}(2\omega,\mathbf{q}) = 0,$ (25)

$$E_{z'}(2\omega,\mathbf{q}) = A\{(p_{zyyxy}\cos^2\phi + p_{zxxxy}\sin^2\phi)u_{xy}(\mathbf{q})$$
$$+[\chi_{zxy}^{(2,0)}f(\mathbf{q})+p_{zxyyy}u_{yy}(\mathbf{q})]\sin 2\phi\}E_s^2(\omega);$$
(26)

(iv) $p(\omega) \rightarrow p(2\omega)$:

$$E_{x'}(2\omega,\mathbf{q}) = A\{2(p_{xxzxy}\cos^2\phi + p_{yyzxy}\sin^2\phi)u_{xy}(\mathbf{q})$$
$$+[2\chi_{zxy}^{(2,0)}f(\mathbf{q})+p_{xyzxx}u_{xx}(\mathbf{q})+p_{yxzyy}u_{yy}(\mathbf{q})]$$
$$\times \sin 2\phi\}E_{x'}(\omega)E_{z'}(\omega),$$
(27)

$$E_{z'}(2\omega,\mathbf{q}) = A\{(p_{zyyxy}\cos^2\phi - p_{zxxxy}\sin^2\phi)u_{xy}(\mathbf{q})$$
$$-[\chi_{zxy}^{(2,0)}f(\mathbf{q})+p_{zxyxx}u_{xx}(\mathbf{q})]\sin 2\phi\}E_{z'}^2(\omega).$$
(28)

The Fourier transform of the dislocation strain tensor components $u_{lm}(\mathbf{q})$ and factor $f(\mathbf{q})$ are determined as

$$u_{lm}(\mathbf{q}) = \frac{1}{V}\int_V u_{lm}(\mathbf{r})\exp(i\mathbf{q}\cdot\mathbf{r})d\mathbf{r},$$
(29)

$$f(\mathbf{q}) = \frac{1}{V}\int_V \exp(i\mathbf{q}\cdot\mathbf{r})d\mathbf{r} = 2\frac{J_1(q_\perp R)}{q_\perp R}\operatorname{sinc}\left(\frac{q_z h}{2}\right),$$
(30)

where $J_1(x)$ is the first-order Bessel function, R is the diameter of the laser spot, $q_\perp$ is the in-plane component of the scattering wave vector $\mathbf{q}$, $\operatorname{sinc}(x) = \sin(x)/x$, and h is the thickness of the crystal. As follows from Eqs. (23)–(28), for different mutual orientations of the beam and crystal coordinate systems, the different components of the nonlinear photoelastic tensor in combination with the second-order NOS tensor will contribute to the HRLS. For example, in the case of $p(\omega) \rightarrow s(2\omega)$ geometry from Eq. (24), we ascertain that for $\phi = 0$ the HRLS intensity is determined by both $\chi^{(2,0)}$ and p_{ijklm} tensors, while for $\phi = \pi/4$ it is determined only by nonzero components of the nonlinear photoelastic tensor. For $s(\omega) \rightarrow p(2\omega)$ and $p(\omega) \rightarrow p(2\omega)$ scattering geometries, from Eqs. (25) and (26) it follows that for $\phi = 0$ the HRLS intensity is determined only by the components of the nonlinear photoelastic tensor, whereas for $\phi = \pi/4$ both tensors contribute to the HRLS. This rotational anisotropy of the HRLS allows us to determine the nonzero components of the nonlinear photoelastic tensor. The angular dependence of the HRLS is also determined by the Fourier transform of the dislocation strain and the factor $f(\mathbf{q})$. Substituting Eqs. (18)–(20) into Eq. (22), we obtain expressions for the Fou-
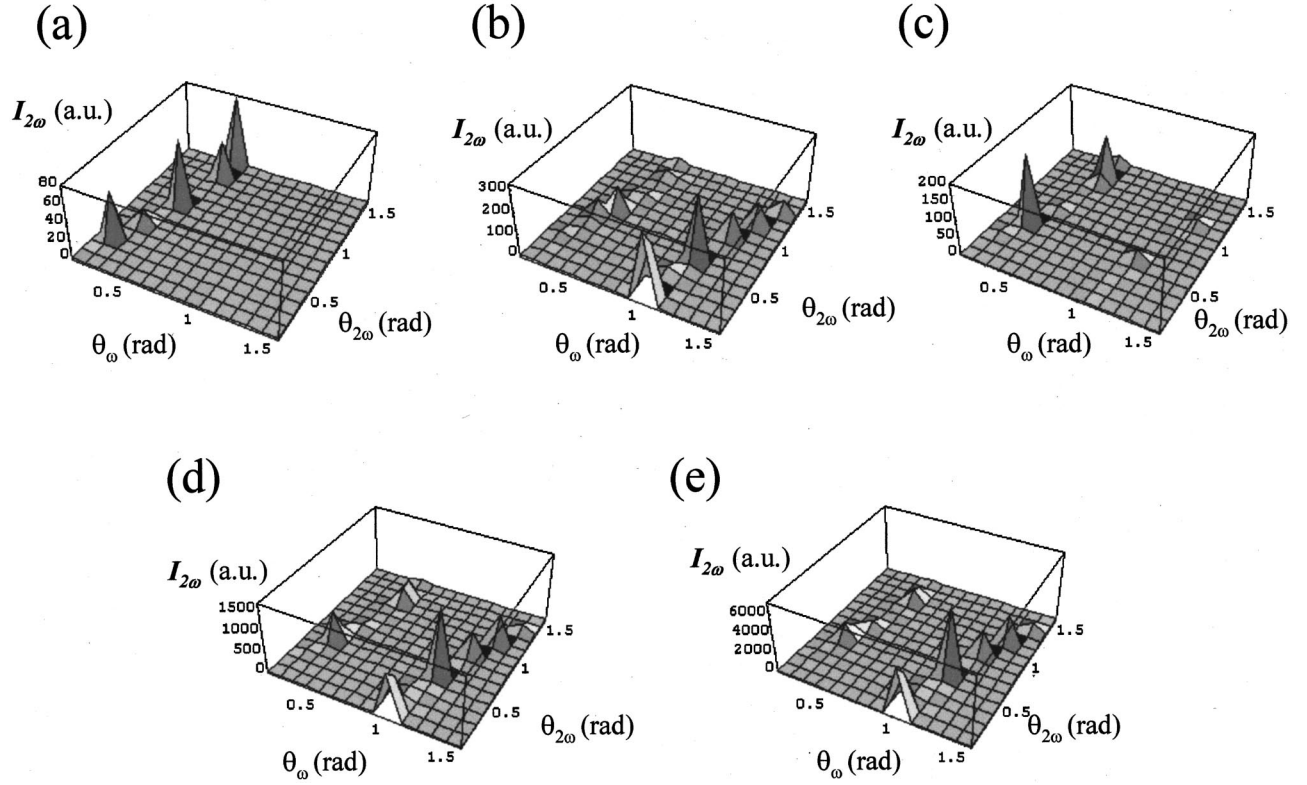
FIG. 3. Angular dependencies of HRLS from a single-edge dislocation for different values of angle ϕ: (a) $\phi = 0$, (b) $\phi = \pi/6$, (c) $\phi = \pi/4$, (d) $\phi = \pi/3$, and (e) $\phi = \pi/2$. (θ_ω and $\theta_{2\omega}$ are the incident angle of fundamental light and the scattered angle of double-frequency light, respectively.)

rier transform of the dislocation strain which determine the electric field of scattered radiation at the second-harmonic frequency in Eqs. (23)–(28):

$$u_{xx}(\mathbf{q}) = \frac{bq_y}{2\pi R^2 q^4 (\nu-1)} \{ q_x^2 [(3+2\nu)(J_0(q_\perp R) - J_0(q_\perp r_d)) + 6(J_1(q_\perp R) - J_1(q_\perp r_d))] + q_y^2 [(2\nu-3)(J_0(q_\perp R) - J_0(q_\perp r_d)) - 2(J_1(q_\perp R) - J_1(q_\perp r_d))] \} \operatorname{sinc}\left(\frac{q_z h}{2}\right),$$

$$(31)$$

$$u_{yy}(\mathbf{q}) = \frac{bq_y}{2\pi R^2 q^4 (\nu-1)} \{ q_x^2 [(2\nu-5)(J_0(q_\perp R) - J_0(q_\perp r_d)) - 6(J_1(q_\perp R) - J_1(q_\perp r_d))] + q_y^2 [(2\nu+1)(J_0(q_\perp R) - J_0(q_\perp r_d)) + 2(J_1(q_\perp R) - J_1(q_\perp r_d))] \} \operatorname{sinc}\left(\frac{q_z h}{2}\right),$$

$$(32)$$

$$u_{xx}(q) = \frac{bq_x}{2\pi R^2 q^4 (\nu-1)} \{ q_x^2 [J_0(q_\perp R) - J_0(q_\perp r_d) + 2(J_1(q_\perp R) - J_1(q_\perp r_d))] - q_y^2 [5 + 6(J_1(q_\perp R) - J_1(q_\perp r_d))] \} \operatorname{sinc}\left(\frac{q_z h}{2}\right),$$

$$(33)$$

where $J_0(x)$ is the zero-order Bessel function, and r_d is the radius of the dislocation core. The HRLS intensity can be calculated using Eqs. (23)–(28) in the standard way. The angular dependence (indicatrix) of the HRLS intensity for the $p(\omega) \rightarrow s(2\omega)$ scattering geometry is shown in Fig. 3 for different values of angle ϕ: $\phi = 0$, $\phi = \pi/6$, $\phi = \pi/4$, $\phi = \pi/3$, and $\phi = \pi/2$. In a similar way, using Eqs. (25), (28), (30), and (31)–(33), it is possible to plot the indicatrix of HRLS for $s(\omega) \rightarrow p(2\omega)$ and $p(\omega) \rightarrow p(2\omega)$ scattering geometries. As follows from Fig. 3, it is possible to find the optimal values of the incident angle θ_ω and scattering angle $\theta_{2\omega}$, to observe the HRLS, and to determine the components of the nonlinear photoelastic tensor.

IV. CONCLUSIONS

In conclusion, the strain-induced changes in the second-order NOS tensor have been phenomenologically investigated by introducing the nonlinear photoelastic tensor. It was shown that the change of symmetry and related strain components can be determined from measurements of angular dependencies of SHG. In addition, we applied nonlinear photoelastic interaction for the description of HRLS by single-edge dislocation. In a similar way, it is possible to investigate HRLS by a screw dislocation. The present phenomenological theory of HRLS by a single dislocation is valid for crystals with a low dislocation density. In the case of a high dislocation density, it is necessary to take into account the distribution of dislocations, their shape (for example, dislocation loops), and mutual orientation.

We believe that the methods of strain-induced nonlinear

optics described in this paper are also very useful for the investigation of other kinds of defect structures in crystals and thin solid films on substrates, because such systems are characterized by strain of different origins induced by lattice mismatch and misfit dislocations located near the film-substrate interfaces.[4,21] We would like to note recent publications[35,36] where enhancement of the SHG signal in a $BaTiO_3/SrTiO_3$ superlattice was observed. This SHG enhancement was qualitatively explained with the large strain induced by lattice mismatch located near the interfaces in a superlattice. The similar experimental results of misfit strain induced enhancement of SHG in pseudocubic $Ba_{0.48}Sr_{0.52}TiO_3$ thin films on the MgO(001) substrates was published in Ref. 37. A theoretical model of misfit strain and misfit dislocation induced SHG, using a nonlinear photoelastic interaction similar to the phenomenological theory of the strain-induced three-photon effects developed above, was proposed in Ref. 38.

In our opinion there are very interesting and promising possibilities to study strain-induced SHG and HRLS in ordered structures such as magnetic crystals and films. Due to magnetoelastic interactions dislocations and strains lead to the formation or so-called dislocation- and strain-induced magnetic domains.[39,40] We hope that these domains can be observed via magnetization-induced SHG, because the nonlinear optical technique is very sensitive for the observation of peculiarities of magnetic domains and domain walls.[41,42]

On the other hand, it should be noted that strain-induced effects can also be observed with nonlinear optical spectroscopy. This is connected with strain-induced changes in the frequency dependencies of the NOS tensors.[10–12,14,16,19] First, the resonance frequency of a SHG signal induced by surface (or interface) strain shifts relative to the bulk resonance frequencies. Second, strain leads to the broadening of corresponding resonant lines at the second-harmonic frequency.[17]

ACKNOWLEDGMENTS

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