Second-harmonic generation from realistic film–substrate interfaces: The effects of strain

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The optical second-harmonic generation from a thin crystalline film on a substrate is theoretically investigated for both s and p polarized incident light. The contributions of lattice misfit strain as well as of misfit dislocation strain to the second-order nonlinear optical susceptibility are described using a nonlinear photoelastic tensor and can be separated by a polarization analysis of the scattered light at the second harmonic frequency. For the s(ω)→s(2ω) and p(ω)→s(2ω) scattering geometries, the nonlinear optical signal will be determined by dislocation strain only, whereas for the s(ω)→p(2ω) and p(ω)→p(2ω) geometries both lattice misfit strain and misfit dislocation strain will contribute. © 2000 American Institute of Physics.

Nonlinear optical methods, such as optical second-harmonic generation (SHG), are very sensitive to the characteristics of surface and interface structures and have therefore found wide application for the investigation of thin solid films on substrates as well as of multilayered structures and superlattices.1,2

In epitaxially grown thin film structures elastic strain appears, localized near the interface and induced by a misfit of the lattice constants of film and substrate.3 The thickness of such a strained layer can vary within a wide range (from a few to a few tens of nanometers)3 and is determined by the elastic parameters of both materials and the misfit f:

\[ f = \frac{a_f - a_s}{a_s}. \]  

In Eq. (1) \( a_f \) and \( a_s \) are the lattice constants of film and substrate, respectively. Such a strain can lead to an extra contribution to the nonlinear optical polarization and has been observed via SHG.4 Moreover, when a film thickness exceeds the critical thickness \( h_c \), misfit dislocations will appear3 that will also contribute to the nonlinear polarization. The influence of such strains and defects on SHG was studied both theoretically and experimentally for semiconductor films and superlattices.4–10 The nonzero components of the second-order nonlinear optical susceptibility tensor \( \chi^{(2)} \) were calculated in a semimicroscopic approach, based on strain-induced changes of the bonds (orbitals) between atoms near the film interfaces.5,6,8 Kulyuk et al.7 mentioned the additional contribution of strain to the nonlinear polarization, however, without giving any explicit derivations. Alternatively, Bottomley9 described the influence of dislocation strain on SHG phenomenologically via a periodic modulation of \( \chi^{(2)} \) due to displacements of the Bravais lattice sites from their positions in the dislocation-free crystal.5 For example, in his description,9 contribution along a screw axis dislocation vanishes because the local average symmetry becomes centrosymmetric. This always results in a decrease of the nonlinear optical response, in contrast to experimental observations where strain leads to the appearance of SHG.11 In this letter, we propose a more general phenomenological description of SHG from an epitaxially grown film on a substrate, taking into account lattice misfit as well as dislocation strain, by using a nonlinear photoelastic tensor similar to Rayleigh light scattering by dislocation strain.12

Consider the interface between a thin crystalline film with cubic symmetry on a thick cubic substrate that is located in the XY plane with the Z axis perpendicular to this interface (see Fig. 1). A thin film of centrosymmetrical (bulk) material does not contain inversion as an element of the point group symmetry, i.e., the symmetry class of a thin film is determined by its orientation.6,11 For example, a thin film of cubic material (point symmetry \( O_h \)) with a four-fold axis \([001]\) directed perpendicularly to the film, is characterized by the point symmetry \( C_{4d} \). We investigate a crystal film with thickness \( t_f \) when \( t_f > h_c \) and misfit dislocations near the interface. The spacing between misfit dislocations \( D_{md} \) is determined by the expression3

\[ D_{md} = \frac{a_f}{f}. \]  

The set of equidistant straight edge dislocations oriented along the Y axis and Burgers vectors parallel to the Y axis is

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characterized by the following nonzero components of the stress tensor: \( \sigma_{yy}^{\text{disl}}(r) \), \( \sigma_{zz}^{\text{disl}}(r) \), and \( \sigma_{yz}^{\text{disl}}(r) \). The strain tensor in the film can be presented as follows:

\[
u_{lm}(r) = u_{lm}^{\text{mish}} \theta(h_c - z) + u_{lm}^{\text{disl}}(r) \theta(z - h_c).
\]

(3)

where \( \theta(z) \) is the Heaviside step function. The first term in Eq. (3) corresponds to the contribution of the lattice misfit strain and the second one describes the dislocation strain. If the film thickness \( t_f \) does not exceed the critical thickness \( h_c \), i.e., \( t_f < h_c \), the film will be free from dislocations and only the first term in Eq. (3) describes the strain in the film. In this case the epilayer will be in a strained state (compressive if \( a_f > a_s \), or tensile if \( a_f < a_s \)) which is characterized by the biaxial tetragonal strain determined by the expression\(^1\)

\[
u_{lm}^{\text{mish}} = f \delta_{lm}.
\]

(4)

where \( \delta_{lm} \) is the two-dimensional Kronecker delta symbol (in our case \( l,m = x,y \)). The dislocation strain can be determined using Hooke’s law,\(^1\) and the corresponding strain tensor is characterized by the following nonzero components:

\[
u_{xy}^{\text{disl}}(r) = -\nu \left( \sigma_{xy}^{\text{disl}}(r) + \sigma_{zz}^{\text{disl}}(r) \right),
\]

\[
u_{yy}^{\text{disl}}(r) = \frac{\sigma_{yy}^{\text{disl}}(r) - \nu \sigma_{zz}^{\text{disl}}(r)}{E},
\]

\[
u_{zz}^{\text{disl}}(r) = \frac{\sigma_{zz}^{\text{disl}}(r) - \nu \sigma_{yy}^{\text{disl}}(r)}{E},
\]

(5)

where \( \nu \) is Poisson’s coefficient and \( E \) is Young’s modulus.\(^1\)

The electric field at the second harmonic frequency of the incident radiation is determined as a solution of the wave equation with the nonlinear polarization \( P_{\text{NL}}^{(2)}(\omega) \) as a source term. In the dipole approximation the latter can be written in the well known form\(^1\)

\[
S_f = \frac{1}{V} \int_V \int E_{\text{inc}}(\omega) \psi(x) \exp(iq \cdot x) dx.
\]

(6)

where the nonlinear polarization has the following form:

\[
\chi^{(2)}_{jk}(\mathbf{r}) = \chi^{(2)}_{jk} + p_{ijklm} u_{lm}(\mathbf{r}).
\]

(7)

Here, \( p_{ijklm} \) and \( u_{lm} \) are the nonlinear photoelastic and strain tensors, respectively. Within the slowly varying amplitude approximation the wave equation for the second harmonic electric field can be written as

\[
2ik_{2\omega} |\mathbf{E}(2\omega, \mathbf{q})| = -\frac{\omega^2}{c^2} \chi^{(2)}_{jk}(\mathbf{r}) E_j(\omega) E_k(\omega) \exp(iq \cdot \mathbf{r}),
\]

(8)

where \( \mathbf{q} = 2k_{\omega} - k_{2\omega} \) is the phase (wave vector) mismatch and \( k_{\omega} \) and \( k_{2\omega} \) are the wave vectors of the fundamental and second harmonic light, respectively. Using the infinite plane wave approximation,\(^1\) we obtain from Eq. (8)

\[
E_i(2\omega, \mathbf{q}) = A \frac{1}{V} \int_V \chi^{(2)}_{ijkl}(\mathbf{r}) E_j(\omega) E_k(\omega) \exp(iq \cdot \mathbf{r}) d\mathbf{r},
\]

(9)

The integral in Eq. (9) is taken over the interaction volume \( V \) and \( n_{2\omega} \) is the refractive index of the film at the second harmonic frequency.

Let us investigate the polarization of light generated at the double frequency for the wave vectors of the incident and reflected light in the XZ plane (see Fig. 1). For the symmetry class \( C_{4v} \), the second-order optical susceptibility \( \chi_{2\omega}^{(2)}(\mathbf{r}) \) (polar third rank) and nonlinear photoelastic \( p_{ijklm} \) (polar fifth rank) tensors are characterized by 41 and 31 independent components, respectively. Using the nonzero components of the \( u_{lm} \), \( \chi_{2\omega}^{(2)} \), and \( p_{ijklm} \) tensors we can now find the contributions of the misfit strain and dislocation strain to the SHG for \( s \) and \( p \) polarizations.

(1) \( s \)-polarized incident light, i.e., \( \mathbf{E}(\omega) = \{0, E_z(\omega), 0\} \):

\[
E_i(2\omega, \mathbf{q}) = A \frac{1}{V} \int_V \chi^{(2)}_{ijk}(\mathbf{r}) E_j(\omega) E_k(\omega) \exp(iq \cdot \mathbf{r}) d\mathbf{r},
\]

(10a)

\[
E_i(2\omega, \mathbf{q}) = A \left[ \chi_{2\omega}^{(2)} \psi(x) \right] E_j(\omega) + p_{2\omega}^{\psi} E_j(\omega)
\]

(10b)

\[
\psi(x) = \frac{1}{V} \int_V \int \psi(x) \exp(iq \cdot x) dx.
\]

(10c)

where \( f(q) \) is a function of the wave vector \( q \). The equations (10a–c) show that the \( s \)-polarized electric field Eq. (10b) at the second harmonic frequency is purely determined by the contribution of misfit dislocations. In contrast, the \( p \)-polarized SHG is determined both by the misfit strain and dislocation strain terms as well as a strain-independent part (10c).

(2) \( p \)-polarized incident light, i.e., \( \mathbf{E}(\omega) = \{ E_x(\omega), 0, E_z(\omega) \} \):

\[
E_i(2\omega, \mathbf{q}) = A \frac{1}{V} \int_V u_{lm}(\mathbf{r}) \exp(iq \cdot \mathbf{r}) d\mathbf{r}.
\]

(11)

Equations (10a–c) show that the \( s \)-polarized electric field Eq. (10b) at the second harmonic frequency is purely determined by the contribution of misfit dislocations. In contrast, the \( p \)-polarized SHG is determined both by the misfit strain and dislocation strain terms as well as a strain-independent part (10c).
In Eq. (12c) we have taken into account that $p_{zzzz} = p_{zzzz}$. Equations (12a–c) show that for $p$-polarized input, the $s$-polarized second-harmonic radiation is determined by strain-induced dislocation strain only, whereas for both $l$-polarized input the $s$-polarized second-harmonic radiation is determined by strain in a epilayer. Then, evaluation of equidistant misfit dislocations the strain-dependent part of tensors were comparable: $u_{xx}(q)\, u_{xy}(q)$, whereas misfit dislocations contribute to the SHG via terms proportional to $u_{xx}(q)$, $u_{yy}(q)$, $u_{zz}(q)$, and $u_{zz}(q)$. As has been shown in numerous recent publications, SHG is a very sensitive method for the investigation of these magnetic films and interfaces, for example, the review article in Ref. 21. In particular the linear magnetization dependence of the SHG signal from magnetic garnet films recently reported by Pavlov et al., was explained as a result of an interference of crystalline and magnetization-induced terms of the nonlinear optical polarization in these thin films. Such a magnetic–nonmagnetic structure of yttrium–iron garnet grown on a gadolinium–gallium garnet substrate is characterized by a small value of the lattice misfit parameter and a large critical thickness. This leads to a deformed layer near the interface, which also contributes to the nonmagnetic part of the $\chi^{(2)}$ tensor, as was observed in Ref. 22.

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