Transient grating diffraction from an interface between two materials: theory and experimental application

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Received July 21, 1992; revised manuscript received January 7, 1993

A general theory for transient grating diffraction from both sides of a single interface is developed. The grating is assumed to have an arbitrary amplitude on both sides of a bound interface with the grating wave vector parallel to the interface. The specific spatial origin of the diffracted light is described for both the reflected and the transmitted geometries of diffraction. In the reflected geometry the signal is dominated by diffraction from a region near the interface that is a small fraction of an optical wavelength wide. The transmitted geometry signal measures primarily the mean bulk properties of the sample. An experimental example of thermal-transient grating diffraction from a $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{SrTiO}_3$ interface is presented. Model calculations for the experimental data are performed to demonstrate that the approach can be used to monitor spatially nonuniform transient grating temporal relaxation.

1. INTRODUCTION

Transient holographic grating spectroscopy has been used over the past decade to study an extensive set of material properties in a wide variety of systems.^{1,2} A transient grating is formed when two coherent light pulses cross at an angle to make an interference pattern in a sample. The light beams then interact with the sample through absorption, electrostriction, or any other mechanism that perturbs the complex index of refraction of the material, leaving a holographic index grating in the sample. A probe pulse, which is temporally delayed relative to the grating excitation pulses, is incident upon the grating. The probe pulse is then diffracted off the holographic index grating in the material to monitor the grating relaxation dynamics. For example, studies of condensed phase systems have deduced the polariton diffusion constants in organic crystals,³ the velocities of flowing liquids *in situ*,⁴ the triplet quantum yields and relaxation dynamics in organic molecules,⁵ the elastic constants of mineral extracts,⁶ the thermal diffusivity constants in thin films,⁷ and the electronic dynamics of rare-earth ions in insulating crystals.⁸ Experiments on semiconductor materials have included ion dose imaging in Si,⁹ surface thermal expansion rates in GaAs,¹⁰ carrier dynamics in GaAs,¹¹ and electronic transport rates in GaAs-AlGaAs multiple quantum wells.^{12,13} Gas phase grating studies in flames have led to the measurement of radical ion concentrations¹⁴ and collisional diffusion constants.¹⁵ Transient gratings have also been used to measure rotational diffusion rates in various liquids^{16,17} and liquid crystals.¹⁸ In addition, the time-domain dynamics of optical phonons¹⁹ and vibrational relaxation rates in proteins²⁰ have been determined.

Early theoretical treatments of transient gratings²¹ were developed by direct analogy with diffraction from transparent volume holograms,^{22,23} where the slowly varying

envelope approximation was used. These treatments considered only bulk diffraction, where the effect of surface interactions were excluded. Previous experimental investigations on transient gratings to study surfaces have concentrated on material-vacuum or material-gas surfaces.^{11,24-27} This work demonstrated that a more detailed theoretical development of transient grating diffraction was needed in which the effects of a vacuum surface would be specifically included. This theoretical development is presented in Refs. 28 and 29, along with experimental results that demonstrate the difference in the surface and bulk contributions to a transient grating signal (see Appendix A). In the transmission geometry there is a signal from both the surface and the bulk. Since the signals are degenerate in both time and propagation direction, they cannot be distinguished from each other. For a sample that is thick compared with a probe wavelength, the signal in the transmission geometry is dominated by the bulk contribution. However, when one considers the reflection or backscattered signal geometry, a significantly different picture emerges. Owing to phase-matching considerations, no reflection signal is generated in the bulk of the material. The surface, however, produces a reflection signal that arises from the boundary conditions for electromagnetic waves. This paper extends the previously developed formalism for transient grating diffraction from a vacuum surface to include the effects that a bound holographic interface (i.e., solid-solid, solid-liquid, liquid-gas, etc.), with significant grating amplitude on both sides of the interface, has on the diffraction efficiency. The formalism is developed in detail for both the transmission and reflection geometries for a single interface between two materials that are assumed to extend to infinity. While real samples have additional interfaces, we have specifically considered the case in which the grating exists only at a single interface in the middle of a layered structure. If the effects of ad-

ditional interfaces or surfaces are to be considered, the results obtained here can be extended by means of more general theoretical approaches that have been developed for four-wave mixing experiments.³⁰

For the theory developed in this paper, when the sample is thick relative to the probe wavelength, the transmitted signal is primarily generated as the probe passes through the bulk of the sample. The signal field amplitude has contributions that are proportional to the grating amplitude at any point in the sample, regardless of the attenuation by the sample. In the reflection geometry, for weakly absorbed light (low optical density), it is demonstrated that the majority of the signal arises from the interfacial region of the sample, provided that there is a grating modulation of the dielectric constant at the interface and a discontinuity of the unperturbed dielectric constant at the interface. The only limitation to the interface selectivity in this case is the depth of the abrupt discontinuity in the dielectric function and the wavelength of the probe light. In general, for no optical absorption of the probe beam, the depth probed extends to an average distance of $\lambda_{\text{vac}}/4\pi n$, where λ_{vac} is the probe wavelength in vacuum and n is the index of refraction of the material. For strongly absorbed light, where the grating amplitude perpendicular to the sample interface typically changes rapidly relative to the optical wavelength, the depth probed is reduced to approximately half the optical absorption depth, or $\lambda_{\text{vac}}/4\pi n$, whichever is less. The exact functional form of the depth probed is discussed in detail in Section 2. Thus the reflection geometry can provide information on the properties of interfacial regions, in contrast to the transmission geometry, which measures the mean bulk properties.

The interface selectivity of this technique is illustrated with experimental results from a thin $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) film on a transparent SrTiO_3 substrate that is excited and probed with beams incident on the film-substrate interface. These experiments measure the thermal kinetics in the YBCO film excited by the laser pulses; hence the signal decay rates are governed by the thermal diffusivity rates in the film and the substrate. Initially all the heat is localized in the absorbing YBCO film near the film-substrate interface. Heat then flows from the interface into the substrate and into the bulk of the film. After a short period of time (~ 1 ns) a significant temperature rise ($\sim 40\%$ of that in the film) is reached inside the substrate. This effects arises from the similar thermal properties of YBCO and SrTiO_3 .^{7,31-33} The theoretical development presented in Section 2 can handle diffraction from gratings that have significant amplitude on both sides of the film-substrate interface. This is an extension of previous theoretical²⁸ and experimental⁷ work that assumed that the grating had a nonzero amplitude on only one side of the interface. If one includes the time-dependent grating spatial relaxation, good quality quantitative fits to the data result, illustrating that the diffraction theory developed here can be applied to complex spatial grating patterns.

2. THEORY

Consider a single interface with a dielectric constant periodically modulated as shown in Fig. 1, with a grating wave vector parallel to the interfacial plane. The grating

is created by two excitation beams with a parallel polarization and wavelength λ_{exc} (measured in vacuum), incident at angles θ and $-\theta$ with respect to the interface normal in the x - y plane. For mathematical clarity, the sample is assumed to extend to infinity, with an unperturbed dielectric constant ϵ_1 for $x < 0$ and ϵ_2 for $x > 0$. The grating excitation produces a dielectric function that has the form

$$\epsilon(x, y, z) = \epsilon_0[\epsilon_1 + \Delta\epsilon(x)\cos(\beta \cdot \mathbf{z})] \quad (x < 0), \quad (2.1)$$

$$\epsilon(x, y, z) = \epsilon_0[\epsilon_2 + \Delta\epsilon(x)\cos(\beta \cdot \mathbf{z})] \quad (x > 0), \quad (2.2)$$

where ϵ_0 is the permittivity of free space and

$$\beta = \frac{-4\pi\epsilon_1^{1/2} \sin \theta}{\lambda_{\text{exc}}} \quad (2.3)$$

is the grating wave vector that is parallel to the $\hat{z}$ axis. The grating amplitude $\Delta\epsilon(x)$ is arbitrary in magnitude along the x dimension that is parallel to the grating fringe peaks. A probe beam with wave vector $\mathbf{k}$, polarized parallel to the y axis, is incident at an angle θ on the left side of the interface at $x = 0$, as shown in Fig. 1. The probe

Fig. 1. a, Transient grating interfacial geometry for reflected and transmitted diffraction. The wave vectors $\mathbf{k}$, $\mathbf{k}'$, and $\mathbf{k}''$ represent the incident, the refracted, and the reflected fundamental waves, respectively, $\mathbf{k}^{T1}$ and $\mathbf{k}^{T2}$ are the transmitted diffraction wave vectors on the left- and right-hand sides of the interface, respectively, and $\mathbf{k}^R$ is the reflected diffraction wave vector. The grating spacing is denoted FS, and the dielectric constant is ϵ_1 for $x < 0$ and ϵ_2 for $x > 0$. b, Wave-vector matching diagram for transient grating interfacial diffraction geometry in part a. The grating wave vector is β , and Δ is the fundamental beam wave-vector magnitude.

beam produces diffracted fields that propagate in both the forward and the backward directions.

The derivation of the diffraction efficiency closely parallels that of previous work on transient grating diffraction from a surface²⁸ and, at a more basic level, that of the parametric interaction of optical waves at the boundary of a nonlinear medium.³⁴ Both of these treatments, however, were derived for material excitation on only one side of the interface for nonuniform and uniform material excitations, respectively. In this paper we extend the previous treatments and consider transient gratings that have significant amplitudes on both sides of a single interface.

We begin the derivation of the diffracted electric-field strength by substituting the dielectric constant given by Eqs. (2.1)–(2.3) into the wave equation

$$-\nabla^2 \mathbf{E}(\mathbf{r}, t) + \frac{\epsilon(x, y, z)}{c^2} \frac{\partial^2 \mathbf{E}(\mathbf{r}, t)}{\partial t^2} = 0. \quad (2.4)$$

Because all the fields are frequency degenerate, the explicit substitution

$$\mathbf{E}(\mathbf{r}, t) = E(r) \hat{k} \exp(-i\omega t) \quad (2.5)$$

is utilized, where $\hat{k}$ is a unit vector pointing in the direction of propagation. To accommodate a series solution to the diffraction problem, it is useful to substitute a product of a normalized dielectric function $\epsilon_i P(x)$ and an amplitude p_0 for the dielectric constant modulation, $\Delta\epsilon(x)$. This yields

$$\Delta\epsilon(x) = \epsilon_1 p_0 P(x), \quad (2.6)$$

where ϵ_i is equal to ϵ_1 for $x < 0$ and ϵ_2 for $x > 0$. The time-independent electric-field amplitude given in Eq. (2.5) is substituted into the wave equation [Eq. (2.4)] to give

$$\frac{\partial^2 E(r)}{\partial x^2} + \frac{\partial^2 E(r)}{\partial z^2} + \frac{\epsilon_0 \epsilon_i \omega^2}{c^2} [1 + p_0 P(x) \cos(\boldsymbol{\beta} \cdot \mathbf{z})] E(r) = 0. \quad (2.7)$$

In the weak-diffraction limit, the coupling constant amplitude p_0 is small ($p_0 \ll 1$). This leads to simplified solutions of Eq. (2.7), since backcoupling of energy from the diffracted beams into the incident probe beam can be neglected. Hence only first-order diffraction needs to be considered. This is a reasonable assumption, since a typical transient grating experiment is conducted in the weak-diffraction limit (with a diffraction efficiency between 10^{-8} and 10^{-2}) to reduce saturation effects in the material.

The solution of Eq. (2.7) is represented as an expansion in powers of p_0 :

$$E(r) = u_0(x) \exp(i\mathbf{k} \cdot \mathbf{z}) + \frac{1}{2} p_0 u_1(x) \{ \exp[i(\mathbf{k} + \boldsymbol{\beta}) \cdot \mathbf{z}] + \exp[i(\mathbf{k} - \boldsymbol{\beta}) \cdot \mathbf{z}] \} + \dots \quad (x < 0), \quad (2.8)$$

$$E(r) = u_0(x) \exp(i\mathbf{k}' \cdot \mathbf{z}) + \frac{1}{2} p_0 u_1(x) \{ \exp[i(\mathbf{k}' + \boldsymbol{\beta}) \cdot \mathbf{z}] + \exp[i(\mathbf{k}' - \boldsymbol{\beta}) \cdot \mathbf{z}] \} + \dots \quad (x > 0), \quad (2.9)$$

The first term in each expression represents the fundamental, or undiffracted, probe field, and the second term represents the first-order diffracted fields. Second-

and higher-order terms in powers of p_0 have been specifically neglected. Substitution of Eqs. (2.8) and (2.9) into Eq. (2.7) gives

$$\frac{d^2 u_0}{dx^2} + \left(\frac{\omega^2 \epsilon_i}{c^2} - k_z^2 \right) u_0 = 0, \quad (2.10)$$

$$\frac{d^2 u_1}{dx^2} + \left[\frac{\omega^2 \epsilon_i}{c^2} - (k_z + \beta)^2 \right] u_1 = \frac{\omega^2 \epsilon}{c^2} P(x) u_0, \quad (2.11)$$

where the x dependence of the incident probe and diffracted field amplitudes are denoted u_0 and u_1 , respectively. The magnitude of the grating wave vector is β [Eq. (2.3)], and k_z is the magnitude of the z component of $\mathbf{k}$ as defined by

$$k_z = \frac{\omega \sqrt{\epsilon_1}}{c} \sin \theta = \frac{\omega \sqrt{\epsilon_2}}{c} \sin \theta'. \quad (2.12)$$

Reflection and refraction of the fundamental probe wave are determined by Eq. (2.10) and lead to the Fresnel relations.³⁵ Equation (2.11) describes the growth of the diffracted wave amplitude. The right-hand side of Eq. (2.11) is a source term that is proportional to the sum of the fundamental incident and reflected probe electric field amplitudes for $x < 0$ and is proportional to the fundamental refracted field amplitude for $x > 0$. This term leads to a transfer of the fundamental, or undiffracted, wave amplitude into the first diffracted order.

The solution of Eq. (2.10) is

$$u_0 = E_0 [\exp(ik_x x)] + A_R \exp(-ik_x x) \quad (x < 0), \quad (2.13)$$

$$u_0 = E_0 A_T \exp(ik_x' x) \quad (x > 0), \quad (2.14)$$

where A_R and A_T are the Fresnel coefficients³⁵ of the reflected and transmitted waves, respectively, and E_0 is the incident probe electric-field amplitude measured at minus infinity. The x component of the wave vectors $\mathbf{k}$, $\mathbf{k}'$, and $\mathbf{k}''$ of the fundamental incident, reflected, and refracted waves, respectively, are given by

$$k_x^2 = \frac{\epsilon_1 \omega^2}{c^2} - k_z^2, \quad (2.15)$$

$$k_x'^2 = \frac{\epsilon_1 \omega^2}{c^2} - k_z^2, \quad (2.16)$$

$$k_x''^2 = \frac{\epsilon_2 \omega^2}{c^2} - k_z^2. \quad (2.17)$$

A graphic representation of the various wave-vector relationships is shown in Fig. 1. For a transversely polarized incident light field, the boundary conditions for Maxwell's equations require that u_0 and du_0/dx be continuous at $x = 0$. The Fresnel relations

$$A_R = \frac{k_x'' - k_x'}{k_x'' + k_x'}, \quad (2.18)$$

$$A_T = \frac{2k_x''}{k_x'' + k_x'} \quad (2.19)$$

result from these boundary conditions.

If one substitutes Eqs. (2.13) and (2.14) into Eq. (2.11), one obtains expressions that govern the diffracted fields

in both the forward and the backward propagating direction. These are given by

$$\frac{d^2 u_1}{dx^2} + k_x^{*2} u_1 = \frac{\epsilon_1 \omega^2 P(x)}{c^2} [\exp(ik_x x) + A_R \exp(-ik_x' x)] E_0 \quad (x < 0), \quad (2.20)$$

$$\frac{d^2 u_1}{dx^2} + k_x^{*2} u_1 = \frac{\epsilon_2 \omega^2 P(x)}{c^2} A_T \exp(ik_x' x) E_0 \quad (x > 0), \quad (2.21)$$

where k_x^* is the x component of the wave vector of either the transmitted or the reflected diffracted field. The squares of the x -component magnitude for both the reflected (k^R) and the transmitted (k^{T1} and k^{T2} for the left- and right-hand sides of the interface, respectively) diffracted wave vectors are

$$(k_x^R)^2 = (k_x^{T1})^2 = \left[\frac{\epsilon_1 \omega^2}{c^2} - (k_z + \beta)^2 \right] \quad (x < 0), \quad (2.22)$$

$$(k_x^{T2})^2 = \left[\frac{\epsilon_2 \omega^2}{c^2} - (k_z + \beta)^2 \right] \quad (x > 0). \quad (2.23)$$

The Green's function solutions for Eqs. (2.20) and (2.21) for excitation at x' are

$$G^{(-)}(x) = \frac{1}{2ik_x^{T1}} \int_{-\infty}^0 \exp(ik_x^{T1}|x - x'|) \phi(x') \exp(ik_x x') dx' + \frac{A_R}{2ik_x^R} \int_{-\infty}^0 \exp(ik_x^R|x - x'|) \phi(x') \exp(-ik_x' x') dx' \quad (x < 0), \quad (2.24)$$

$$G^{(+)}(x) = \frac{A_T}{2ik_x^{T2}} \int_0^{\infty} \exp(ik_x^{T2}|x - x'|) \phi(x') \exp(ik_x' x') dx' \quad (x > 0) \quad (2.25)$$

for diffraction from the right- and left-hand sides of the interface, denoted (+) and (-), respectively. The coupling parameter

$$\phi(x) = \frac{\epsilon_1 \omega^2 E_0 P(x)}{c^2} \quad (2.26)$$

contains the spatially nonuniform grating amplitude dependence $P(x)$. The diffracted field amplitudes can be written as the sum of the homogeneous and the Green's function (particular) solutions to Eqs. (2.20) and (2.21):

$$u_1^{(-)}(x) = a_R \exp(-ik_x^R x) + G^{(-)}(x) \quad (x < 0), \quad (2.27)$$

$$u_1^{(+)}(x) = a_T \exp(ik_x^T x) + G^{(+)}(x) \quad (x > 0), \quad (2.28)$$

where a_R and a_T are the amplitudes of the reflected and transmitted interface field amplitudes.³⁴ One determines these amplitudes by matching the boundary conditions for the tangential components of the electric and the magnetic fields, which reduce again to

$$u_1^{(-)} = u_1^{(+)}, \quad (2.29)$$

$$\frac{du_1^{(-)}}{dx} = \frac{du_1^{(+)}}{dx} \quad (2.30)$$

at $x = 0$.

To implement the boundary conditions, the Green's functions must be explicitly evaluated at $x = 0$. This dictates that

$$\exp(ik_x^{T1}|x - x'|) = \exp[ik_x^{T1}(x - x')], \quad (2.31)$$

$$\exp(ik_x^R|x - x'|) = \exp[ik_x^R(x - x')] \quad (2.32)$$

for $G^{(-)}$ at $x = 0$ and $x' < 0$ and

$$\exp(ik_x^{T2}|x - x'|) = \exp[ik_x^{T2}(x' - x)] \quad (2.33)$$

for $G^{(+)}$ at $x = 0$ and $x' > 0$. To simplify the solution, the fundamental undiffracted beam is assumed to be incident at the Bragg angle; thus

$$k_x = k_x' = k_x^{T1} = k_x^R \equiv k_{1x}, \quad (2.34)$$

$$k_x' = k_x^{T2} \equiv k_{2x}. \quad (2.35)$$

After substituting Eqs. (2.31)–(2.35) into Eqs. (2.27) and (2.28) and matching the boundary conditions of Eqs. (2.29) and (2.30), one obtains

$$a_R = \frac{2k_{2x}}{k_{1x} + k_{2x}} M^{(+)} + \frac{k_{1x} - k_{2x}}{k_{1x} + k_{2x}} M^{(-)} \quad (2.36)$$

for the reflected interface field amplitudes and

$$a_T = \frac{k_{2x} - k_{1x}}{k_{1x} + k_{2x}} M^{(+)} + \frac{2k_{1x}}{k_{1x} + k_{2x}} M^{(-)} \quad (2.37)$$

for the transmitted interface field amplitudes, where

$$M^{(-)} = \frac{1}{2ik_{1x}} \int_{-\infty}^0 [A_R \exp(-2ik_{1x} x') + 1] \phi(x') dx', \quad (2.38)$$

$$M^{(+)} = \frac{1}{2ik_{2x}} \int_0^{\infty} A_T \exp(2ik_{2x} x') \phi(x') dx'. \quad (2.39)$$

Substitution of Eqs. (2.36)–(2.39) into the expressions for u_1 [Eqs. (2.27) and (2.28)] gives the final expressions for the diffracted field amplitudes. First the Green's functions must be explicitly evaluated at $x = \pm\infty$, which dictates that

$$\exp(ik_{2x}|x - x'|) = \exp[ik_{2x}(x - x')] \quad (2.40)$$

for $G^{(+)}$ at $x = \infty$ and

$$\exp(ik_{1x}|x - x'|) = \exp[ik_{1x}(x' - x)] \quad (2.41)$$

for $G^{(-)}$ at $x = -\infty$. The reflected diffracted field amplitude observed as x approaches $-\infty$ is then given by

$$u_1^R = \frac{-i \exp(-ik_{1x} x)}{2} \left[\frac{A_R}{k_{1x}} \int_{-\infty}^0 \phi(x') dx' + \frac{(A_R)^2}{k_{1x}} \int_{-\infty}^0 \exp(-2ik_{1x} x') \phi(x') dx' + \frac{1}{k_{1x}} \int_{-\infty}^0 \exp(+2ik_{1x} x') \phi(x') dx' + \frac{A_T A_T'}{k_{2x}} \int_0^{+\infty} \exp(+2ik_{2x} x') \phi(x') dx' + \frac{A_R}{k_{1x}} \int_{-\infty}^0 \phi(x') dx' \right] \\ \propto \mathbf{C} + \mathbf{H} + \mathbf{E} + \mathbf{F} + \mathbf{D}, \quad (2.42)$$

where **C**, **H**, **E**, **F**, and **D** represent the five integral terms with their coefficients in order, respectively. The transmitted diffracted field amplitude observed as x approaches $+\infty$ is given by

$$u_1^T = \frac{-iA_T \exp(+ik_{2x}x)}{2} \left[\frac{1}{k_{1x}} \int_{-\infty}^0 \phi(x') dx' + \frac{1}{k_{2x}} \int_0^{+\infty} \phi(x') dx' + \frac{A_R}{k_{1x}} \int_{-\infty}^0 \exp(-2ik_{1x}x') \phi(x') dx' + \frac{-A_R}{k_{2x}} \int_0^{+\infty} \exp(+2ik_{2x}x') \phi(x') dx' \right] \\ \propto \mathbf{A} + \mathbf{B} + \mathbf{G} + \mathbf{I}, \quad (2.43)$$

where **A**, **B**, **G**, and **I** represent the four integral terms with their coefficients in order, respectively. Here,

$$A_R = \frac{k_{1x} - k_{2x}}{k_{1x} + k_{2x}} = \frac{\sqrt{\epsilon_1} \cos \theta - \sqrt{\epsilon_2} \cos \theta'}{\sqrt{\epsilon_1} \cos \theta + \sqrt{\epsilon_2} \cos \theta'}, \quad (2.44)$$

$$A_T = \frac{2k_{1x}}{k_{1x} + k_{2x}} = \frac{2\sqrt{\epsilon_1} \cos \theta}{\sqrt{\epsilon_1} \cos \theta + \sqrt{\epsilon_2} \cos \theta'} \quad (2.45)$$

are the electric-field Fresnel reflection and transmission coefficients for propagation from left to right across the interface and

$$A_T' = \frac{2k_{2x}}{k_{1x} + k_{2x}} = \frac{2\sqrt{\epsilon_2} \cos \theta'}{\sqrt{\epsilon_1} \cos \theta + \sqrt{\epsilon_2} \cos \theta'} \quad (2.46)$$

is the electric-field Fresnel transmission coefficient for propagation from right to left across the interface.

The general expressions for the reflected and the transmitted field amplitudes [relations (2.42) and (2.43)] have nine integral terms (**A**–**I**) that can be qualitatively understood. Each of the nine terms has a specific geometric origin, as shown in Fig. 2. The incident, refracted, and reflected fundamental (undiffracted) beams are denoted by solid lines and are labeled **k**, **k'**, and **k''**, respectively. Four of the diffracted waves (**A**, **B**, **E**, and **F**), shown by dashed arrows in Fig. 2, have amplitudes significantly stronger than the other five diffracted waves, which are drawn as dotted arrows. The five weak waves arise from integral terms in relations (2.42) and (2.43) that have an amplitude proportional to A_R or A_R^2 , where A_R is the electric-field reflection coefficient for the incident fundamental beam **k**. Hence, if the interfacial dielectric constants are approximately equal, then A_R goes to zero and the four strong terms completely dominate the reflected and transmitted signals. However, if the interface arises from two dissimilar materials, such as a metal and an insulator, then all nine terms could have significant contributions.

The first integral term in relation (2.43) is simply the bulk transmitted diffraction that originates from the left-hand side of the interface. The bulk holographic diffraction field builds up linearly with distance and the grating modulation amplitude, as evidenced by the unity amplitude in front of the coupling coefficient $\phi(x')$ inside the first integral. This component, denoted **A** in Fig. 2, originates from diffraction of the incident fundamental beam **k**. The second integral term in relation (2.43) again arises from bulk, or holographic, diffraction in the transmitted direction, but the signal now originates from the

right-hand side of the interface. Field **B** in Fig. 2 represents this term, which originates from the refracted fundamental beam, **k'**. It is important to note that **A** and **B** add in phase and propagate in parallel directions for $x > 0$, where either of these waves would be detected. Hence, if the interfacial discontinuity vanishes (i.e., $\epsilon_1 = \epsilon_2$), then **A** and **B** would be experimentally indistinguishable.

Fields **C** and **D** also build up as bulk (holographic) diffraction fields; however, they propagate in a backscattered geometry parallel to the other reflected diffraction terms. The amplitude of **C**, described by the first integral term in relation (2.42), builds up from the bulk diffraction of **k** as it propagates on the left-hand side of the interface. A simple mirror reflection from the interface of the bulk transmitted signal **A**, such that it propagates in the reflected diffracted direction, gives the field **C**. This field is attenuated over **A** by a factor of the Fresnel reflection coefficient A_R . The reflected fundamental beam **k''** also undergoes bulk (holographic) diffraction as it propagates in the negative direction. This gives rise to the last bulk term **D**, described by the fifth integral term in relation (2.42). Fields **C** and **D** are attenuated, because of reflection from the interface, relative to the two most intense bulk transmitted diffraction terms **A** and **B**. This attenuation would typically be of the order of 1–10% for a dielectric interface between two similar materials.

Fields **E**, **F**, and **G** arise from diffraction in a backscattered (phase-conjugate direction) geometry from spatial nonuniformities in the x dimension of the grating amplitude. The curved arrows in Fig. 2 represent this type of diffraction. Specifically, the dashed arrows **E** and **F** arise from the third and fourth integral terms of relation (2.42), which represent reflected, or backscattered, diffraction that originates from the left- and right-hand sides of the interface, respectively. The exponential functions in the integral terms of relation (2.42) lead to destructive interference of the diffracted wave amplitude, if the grating amplitude is smooth in the x dimension. If the grating amplitude in the bulk of the material is not smooth (i.e., $d\Delta\epsilon/dx \approx |\mathbf{k}|$), then the symmetry of the interference is broken, giving rise to relatively significant diffracted amplitudes from the bulk of the material. The third term

Fig. 2. Transient grating electric-field components for diffraction from an interface. The solid arrows **k**, **k'**, and **k''** represent the incident, refracted, and reflected fundamental waves, respectively. Arrows **A**–**I** represent nine different diffracted field components for reflected diffraction (left-propagating arrows) and transmitted diffraction (right-propagating arrows).

(E) in relation (2.42) has an amplitude proportional to $1/k_{1x}$, while the fourth term (F) has an amplitude proportional to $A_T A_T'/k_{2x}$, making these the two strongest terms for reflected diffracted fields. The $A_T A_T'$ factor in the amplitude of the fourth integral term in relation (2.42) arises because this field component must propagate across the interface from left to right before undergoing diffraction and then must propagate back across the interface before being measured at $x \ll 0$. For most dielectric interfaces this attenuation has negligible strength (i.e., $A_T \approx A_T' \approx 0.9$).

Field G represents backscattered (phase-conjugate direction) diffraction from the reflected fundamental beam $\mathbf{k}''$. This results in a field that propagates in the transmitted diffraction direction. The third term of relation (2.43) represents this process and has an amplitude proportional to A_R/k_{1x} . This term is attenuated both from interference arising from the exponential factor in the integral and from the amplitude factor A_R that arises from the reflection of the fundamental beam. Hence, G has a small amplitude relative to the A and B transmitted diffraction fields.

Fields H and I result solely from matching the boundary conditions at the interface.³⁴ The second integral term in relation (2.42) represents field H in Fig. 2, which contributes to the total reflected diffracted field amplitude. The field strength of H is reduced from both destructive interference in the exponential function and from the small amplitude A_R^2/k_{1x} (10^{-4} – $10^{-2}\%$ for most dielectric interfaces). Consequently, H will usually be overwhelmed by the much more intense fields generated by E and F. The fourth integral term in relation (2.43) is associated with field I in Fig. 2. This term adds to the transmitted diffracted field amplitude but has a relatively small field strength contribution from both the destructive interference in the exponential function of the term and the small amplitude $-A_R/k_{2x}$. As a result, I will normally be eclipsed by the much more intense fields generated from A and B in the transmitted geometry.

Expressions for the diffraction efficiency at the Bragg angle are obtained when one substitutes Eqs. (2.26), (2.45), and (2.46) and relations (2.42) and (2.43) into Eqs. (2.8) and (2.9). The general reflected diffraction efficiency observed as x approaches $-\infty$ is proportional to

$$\begin{aligned} \eta^R = & \left| \frac{E^R}{E_0} \right|^2 \\ & \propto \left(\frac{\omega}{c} \right)^2 \left| \frac{2(\sqrt{\epsilon_1} \cos \theta - \sqrt{\epsilon_2} \cos \theta')}{\sqrt{\epsilon_1} \cos \theta (\sqrt{\epsilon_1} \cos \theta + \sqrt{\epsilon_2} \cos \theta')} \right. \\ & \times \int_{-\infty}^0 \Delta\epsilon(x') dx' + \frac{(\sqrt{\epsilon_1} \cos \theta - \sqrt{\epsilon_2} \cos \theta')^2}{\sqrt{\epsilon_1} \cos \theta (\sqrt{\epsilon_1} \cos \theta + \sqrt{\epsilon_2} \cos \theta')^2} \\ & \times \int_{-\infty}^0 \exp\left(-2i\sqrt{\epsilon_1} \frac{\omega}{c} \cos \theta x'\right) \Delta\epsilon(x') dx' \\ & + \frac{1}{\sqrt{\epsilon_1} \cos \theta} \int_{-\infty}^0 \exp\left(+2i\sqrt{\epsilon_1} \frac{\omega}{c} \cos \theta x'\right) \Delta\epsilon(x') dx' \\ & + \frac{4\sqrt{\epsilon_1} \cos \theta}{(\sqrt{\epsilon_1} \cos \theta + \sqrt{\epsilon_2} \cos \theta')^2} \\ & \times \int_0^{+\infty} \exp\left(+2i\sqrt{\epsilon_2} \frac{\omega}{c} \cos \theta x'\right) \Delta\epsilon(x') dx' \Big|^2. \quad (2.47) \end{aligned}$$

The general transmitted diffraction efficiency observed as x approaches $+\infty$ is proportional to

$$\begin{aligned} \eta^T = & \left| \frac{E^T}{E_0} \right|^2 \\ & \propto \left[\frac{\omega\sqrt{\epsilon_1} \cos \theta}{c(\sqrt{\epsilon_1} \cos \theta + \sqrt{\epsilon_2} \cos \theta')} \right]^2 \left| \frac{1}{\sqrt{\epsilon_1} \cos \theta} \right. \\ & \times \int_{-\infty}^0 \Delta\epsilon(x') dx' + \frac{1}{\sqrt{\epsilon_2} \cos \theta'} \int_0^{+\infty} \Delta\epsilon(x') dx' \\ & + \frac{(\sqrt{\epsilon_1} \cos \theta - \sqrt{\epsilon_2} \cos \theta')}{\sqrt{\epsilon_1} \cos \theta (\sqrt{\epsilon_1} \cos \theta + \sqrt{\epsilon_2} \cos \theta')} \\ & \times \int_{-\infty}^0 \exp\left(-2i\sqrt{\epsilon_1} \frac{\omega}{c} \cos \theta x'\right) \Delta\epsilon(x') dx' \\ & + \frac{(\sqrt{\epsilon_2} \cos \theta' - \sqrt{\epsilon_1} \cos \theta)}{\sqrt{\epsilon_2} \cos \theta' (\sqrt{\epsilon_1} \cos \theta + \sqrt{\epsilon_2} \cos \theta')} \\ & \times \int_0^{+\infty} \exp\left(+2i\sqrt{\epsilon_2} \frac{\omega}{c} \cos \theta x'\right) \Delta\epsilon(x') dx' \Big|^2. \quad (2.48) \end{aligned}$$

ϵ_1 , ϵ_2 , and $\Delta\epsilon(x')$ can all take on complex values owing to absorption of the light field.

Two important limiting cases of the general diffraction efficiency expressions will now be discussed. The first case is that of holographic diffraction from an interface between two materials of virtually identical index of refraction (i.e., $\epsilon_1 \approx \epsilon_2 \approx \epsilon$). For this case, the Fresnel reflection coefficient A_R is approximately zero, and all the interfacial terms in relations (2.42) and (2.43) (C, D, G, H, and I) drop out. This is equivalent to inserting a fictitious, or virtual, interface in the middle of a single material slab. The remaining terms in relation (2.42) and (2.43) (A, B, E, and F) lead to

$$\eta^R \propto \left| \frac{\omega}{c} \int_{-\infty}^{+\infty} \exp\left(+2i\sqrt{\epsilon} \frac{\omega}{c} \cos \theta x'\right) \Delta\epsilon(x') dx' \right|^2 \quad (2.49)$$

for the reflected diffraction efficiency and

$$\eta^T \propto \left| \frac{\omega}{c} \int_{-\infty}^{+\infty} \Delta\epsilon(x') dx' \right|^2 \quad (2.50)$$

for the transmitted diffraction efficiency from a material of infinite thickness (i.e., no interfacial or surface interactions of either the diffracted or fundamental beams). These expressions would refer, for example, to the case of a solid nonabsorbing material surface in contact with an index matched fluid. There would be a real solid-liquid interface that could lead to a variety of mechanical surface phenomena such as surface acoustic wave propagation that would have an abrupt grating amplitude discontinuity at the interface. However, the interface would not exist from an optical perspective. Consequently, the optical surface terms (C, D, G, H, and I) are not present in the diffraction efficiency expressions. In addition to the special case for solid-liquid interfaces described above, reflected diffraction in relation (2.49) could also arise from the irregularities in the grating amplitude perpendicular to the grating wave vector (x axis) inside the bulk of the material. This is similar to the reflection of radio waves from the nonuniformities of the dielectric constant in the

global ionosphere. Hence, the grating diffraction in reflection has a substantial amplitude only whenever there are grating amplitude irregularities with a spatial frequency higher than the wave vector of the probe light. In contrast, transmitted diffraction probes the entire sample uniformly. The signal in the transmitted direction is simply the spatial integral, over average perturbation, in the material, as given by relation (2.50).

The second limiting case is diffraction from a vacuum surface. A vacuum is assumed for $x < 0$ ($\epsilon_1 = 1$), and a material dielectric constant ϵ_2 is assumed for $x > 0$. Since there can be no material perturbation on the vacuum side of the interface, all the integrals from $-\infty$ to 0 in relations (2.47) and (2.48) have an integrand equal to zero (i.e., $\Delta\epsilon(x') \equiv 0$ for $x' < 0$). Thus the reflected diffraction efficiency expression reduces to

$$\eta^R \propto \left| \frac{\omega}{c} \int_0^{+\infty} \exp\left(+2i\sqrt{\epsilon_2} \frac{\omega}{c} \cos \theta' x'\right) \Delta\epsilon(x') dx' \right|^2, \quad (2.51)$$

and the transmitted diffraction efficiency reduces to

$$\begin{aligned} \eta^T \propto & \left| \frac{\omega}{c} \int_0^{+\infty} \Delta\epsilon(x') dx' + \frac{(\sqrt{\epsilon_2} \cos \theta' - \cos \theta)}{(\sqrt{\epsilon_2} \cos \theta' + \cos \theta)} \right. \\ & \times \left. \int_0^{+\infty} \exp\left(+2i\sqrt{\epsilon_2} \frac{\omega}{c} \cos \theta' x'\right) \Delta\epsilon(x') dx' \right|^2. \quad (2.52) \end{aligned}$$

Although this limiting case was discussed in detail previously,²⁸ we summarize some of the important features of vacuum-surface transient holographic diffraction here for completeness.

For samples that are optically thick (much greater than the wavelength of light), the first integral in relation (2.52), which arises from bulk holographic diffraction, dominates in magnitude over the second integral. The second integral is a surface term that originates from matching the boundary conditions at the interface. For weakly absorbing grating excitation beams, where the grating amplitude is typically smooth relative to a wavelength of light, the surface term is generated from a region that is approximately equal to one tenth or less of the wavelength of light inside the material. The reflected diffracted geometry described by relation (2.51) has only one surface integral term. This term selectively probes the surface region (depth $\leq \lambda_{\text{vac}}/(4\pi n)$) that arises from the interference created by the exponential function in the integrand. The transmitted geometry preferentially probes the mean bulk properties, while the reflected geometry selectively samples the properties of the surface region. Therefore, the transient grating technique has the ability to probe differing regions of the same sample selectively by varying the detection geometry.

For a quantitative understanding of the magnitude of the surface selectivity, it is useful to define an effective grating depth for the surface component of the signal. The grating excitation is taken to be uniform on the right-hand side of the interface [i.e., $\Delta\epsilon(x) = \Delta\epsilon_0$ ($x > 0$)], and the grating amplitude is zero on the left-hand side (vacuum side) of the interface ($x < 0$). The incident fundamental beam has a complex (absorptive) wave vector inside the media. This is given by

$$\tilde{k} = (2\pi/\lambda)\tilde{n}, \quad (2.53)$$

where the complex index of refraction $\tilde{n}$ is defined as

$$\tilde{n} = n + i\gamma. \quad (2.54)$$

To define the effective grating surface probe depth x_s , we introduce the second moment in the x dimension of the reflected grating signal:

$$x_s^2 = |\sqrt{\eta^R} x|^2 / \eta^R, \quad (2.55)$$

where η^R is defined in relation (2.51). A Bragg angle of zero degrees corresponding to infinite grating fringe spacing is now assumed for simplification. This retains the essential features necessary for understanding the signal spatial origin without any unnecessary complexity. Thus relation (2.51) is reduced to

$$\eta^R \propto E_R^* E_R = \frac{\omega^2}{c^2} \left| \int_0^{+\infty} \exp(2i\tilde{k}x') \Delta\epsilon_0 dx' \right|^2, \quad (2.56)$$

where $\tilde{k}$ is the wave vector of the refracted probe beam inside the media. It is convenient to define a quantity D as

$$D = \frac{\omega\Delta\epsilon_0}{c} \int_0^{+\infty} \exp(2i\tilde{k}x) x dx, \quad (2.57)$$

such that the effective grating depth x_s from Eq. (2.44) can be rewritten as

$$x_s^2 = \frac{D^* D}{E_R^* E_R}. \quad (2.58)$$

Integration of Eq. (2.57) by parts yields

$$D = \frac{-\omega\Delta\epsilon_0}{c} \int_0^{+\infty} \frac{\exp(2i\tilde{k}x)}{2i\tilde{k}} dx = \frac{-E_R}{2i\tilde{k}}. \quad (2.59)$$

Substitution of Eq. (2.59) into Eq. (2.58) leads to an effective grating depth squared of

$$x_s^2 = \frac{1}{4\tilde{k}^* \tilde{k}}. \quad (2.60)$$

Substituting for the complex probe wave vector

$$\tilde{k} = \frac{2\pi}{\lambda} \tilde{n} \hat{x} = \frac{2\pi}{\lambda} (n + i\gamma) \hat{x}, \quad (2.61)$$

where λ is the probe wavelength measured in vacuum, yields a grating depth of

$$x_s = \frac{\lambda}{4\pi n(1 + (\gamma/n)^2)^{1/2}}. \quad (2.62)$$

The effective probe depth, x_s , is plotted in Fig. 3 versus the imaginary component of the index of refraction, γ . The important point to note is that the deepest effective penetration of the signal origin into the sample is less than or equal to the optical wavelength in the sample divided by 4π . For intensely absorbing samples, such as metals, the depth probed can fall to several percent of a probe wavelength. In the case of a thick sample with a slowly varying index modulation, the transmission case uniformly probes the bulk, while the reflection case probes only the interfacial region with an effective depth given by x_s .

Fig. 3. Effective grating probe depth x_s normalized to the wavelength of the probe beam in the media versus the ratio of the imaginary to the real component of the index of refraction, γ/n . For no absorption, i.e., $\gamma/n = 0$, the depth probed is less than 8% of the probe wavelength in the material. For absorptive materials this distance is reduced.

For the general case of diffraction from both sides of an interface, there are multiple terms [i.e., terms **E**, **F**, **G**, **H**, and **I** in relations (2.42) and (2.43)] arising from the interface that have equivalent probe depths for uniform grating excitation. These terms have unequal amplitudes and different probe depths on either side of the interface arising from the differing index of refraction. The net result is that a depth of $\lambda_{\text{vac}}/(4\pi n)$ or less is probed on each side of the interface. This leads to a powerful approach for observing both interfacial and bulk phenomena with the same transient grating experimental method.

3. EXPERIMENTAL EXAMPLE

Experimental results and model calculations are presented in this section to demonstrate transient grating diffraction from a solid-solid interface in both the transmitted and the reflected diffraction geometries. The results presented here will be expanded in scope and the material science implications will be discussed elsewhere.³⁶ Here we are interested in the interfacial transient grating theoretical theory. A thermal transient grating is excited in a thin YBCO film at the interface between the YBCO and an optically transparent $\text{SrTiO}_3(100)$ substrate. One detects the grating by diffracting a temporally delayed probe beam from the spatial modulations of the dielectric constant. The experimental results demonstrate the differences between the reflection and transmission diffraction geometries. If one monitors the grating diffraction in both the reflected and transmitted geometries, additional information is extracted that cannot be obtained from any single diffraction geometry.

A. Experimental Procedures

The output of a *Q*-switched, mode-locked Nd:YAG laser is frequency doubled to a wavelength of 532 nm and used to drive a sync-pumped dye laser at a wavelength of 561 nm. The dye laser is cavity dumped to produce ~ 50 -ps, 20- μJ pulses at a 1-kHz repetition rate. Single dye pulses are attenuated and then split into three beams. Two beams

are focused to a 200- μm -diameter spot size and crossed at an angle 2θ upon the interface between a thin (190-nm) film of optically absorbing YBCO and an optically transparent $\text{SrTiO}_3(100)$ insulating substrate, as shown in Fig. 4. Both excitation beams are incident from the substrate side of the interface. This produces a sinusoidal interference pattern in the YBCO and generates a grating parallel to the interface, as shown in Figs. 1 and 4. Absorption of the light produces excited electrons that rapidly relax³⁷ (~ 1 ps), raising the phonon bath temperature and thus producing a sinusoidal heat pattern in the thin sample. The generation of the thermal grating (or dielectric constant modulation) leads to a density grating, caused by thermal expansion of the YBCO film and the excitation of acoustic waveguide modes^{26,38,39} (the oscillations in Fig. 5). Both heat and acoustic waves modify the film's density in a spatially periodic manner. The density modulations result in changes in the sample's complex index of refraction.²¹ The modulation of the imaginary part of the index of refraction in the YBCO film is approximately equal to the modulation of the real part of the index of refraction in the film. Heat flowing into the substrate can also produce a signal by modulating the real part of the transparent substrate's index of refraction.

The third pulse, which is the probe, is incident at the Bragg angle from the same side of the sample as the excitation beams. The probe is temporally delayed and diffracted from the transient holographic grating in both transmitted and reflected geometries to monitor the decay of the grating, as depicted in Fig. 1. The diffracted signal is detected with a cooled photomultiplier tube and gated integrator. To improve the signal-to-noise ratio, a computer is used to signal average multiple scans of the

Fig. 4. Transient grating experimental excitation geometry. Grating excitation beams pass through the transparent SrTiO_3 substrate and excite a grating only in the absorbing YBCO film. Shortly after this excitation (~ 1 ns) the heat that is deposited in the film flows into the substrate, leaving a thermal grating in both the film and the substrate. A probe beam that is approximately collinear with one of the excitation beams is incident at the Bragg angle from the substrate side of the sample.

Fig. 5. Transient grating thermal signal from a YBCO–SrTiO₃ interface, as shown in Fig. 4, in both the reflected and the transmitted geometries of diffraction with theoretical calculations.

delay line. In addition, the excitation and probe beams are orthogonally polarized, and the signal is then detected with an analyzing polarizer parallel to the incident probe polarization. This filters out scattered light from the excitation beams and improves the signal-to-noise ratio by ~ 1 order of magnitude.

The transient grating experiments are performed in a vacuum chamber (10^{-6} Torr) on *c*-axis-oriented thin films of YBCO (190 nm thick) grown on SrTiO₃(100) substrates. The optical density of the YBCO films at the experimental wavelength is 0.9. A detailed description of the sample characteristics and preparation techniques is provided in the literature.^{40,41} The grating wave vector is parallel to the film interface which defines the *a*–*b* plane of the sample (*y*–*z* plane in Fig. 4). Experiments are performed at room temperature with a grating fringe spacing of 4.4 μm .

B. Model Calculation

The signal is calculated for the different probe diffraction geometries shown in Fig. 1. Only diffusional heat flow will be considered, i.e., the mean-free path of the phonons is assumed to be short compared with the sample thickness. We calculate the time-dependent transient grating diffraction efficiency from the final result given in relations (2.47) and (2.48). The grating amplitude $\Delta\epsilon$ is proportional to the time-dependent grating peak-to-null temperature $\Delta T(x, t)$,^{7,21}

$$\Delta\epsilon(x, t) \propto \Delta T(x, t), \quad (3.1)$$

where $\Delta T(x, t)$ is obtained from the numerical solutions to the diffusion equation

$$D\nabla^2 T = \frac{\partial T}{\partial t}. \quad (3.2)$$

The complex-valued proportionality constant in relation (3.1) contains the complex index of refraction and the elasto-optic constant; the parameters are included in the model calculation but will not be discussed here for brevity.⁷ This numerical solution for the dielectric constant modulation amplitude is then substituted into relations (2.47) and (2.48) for the diffraction efficiency in the reflected and transmitted geometries of diffraction, respec-

tively. The dielectric constants used in the calculations are obtained from a Kramers–Kronig analysis of the reflection and transmission coefficients from the film and the substrate. This gives indices of refraction of 1.5 for SrTiO₃ and $1.7 + i0.49$ for YBCO.

The diffusion equation [Eq. (3.2)] is solved numerically by means of the Crank–Nicolson method⁴² with the appropriate boundary and initial conditions. The vacuum surface of the 190-nm thick YBCO film and the vacuum surface of the substrate are assumed to have a reflecting boundary condition, i.e., $dT/dx = 0$ at $x = 190$ nm and $x = 3000$ nm. An initial condition of

$$T(x, y, z, t = 0) = T_0/2 \exp(-\Omega x)[1 + \cos(\beta t)], \quad (x > 0) \quad (3.3)$$

$$T(x, y, z, t = 0) = 0 \quad (x < 0) \quad (3.4)$$

is used, where β is the grating wave vector and T_0 is the initial temperature jump at the incident interface. The exponential term in Eq. (3.3) accounts for Beer's law absorption of the excitation beams. Literature values for the thermal diffusivity D are used for SrTiO₃ (Refs. 32 and 33) and YBCO (Ref. 7) in the *c*-axis direction. The three-dimensional problem can be simplified if one assumes that diffusion of heat between the grating fringes and across the laser spot size is negligible. This is a reasonable assumption, since the grating fringe spacing (4.4 μm) and the laser spot size (~ 200 μm) are very large compared with the mean thermal diffusional distance (~ 100 nm) reached on the experimental time scale of 10 ns.

Phonon propagation across the interfacial region is assumed to be perfectly diffusive, i.e., every phonon that reaches the interface has a 50% probability of propagating left or right. The equal probability for propagating in either direction is an important point that is not always valid for interfaces that have a large thermal boundary resistance. This is discussed in detail elsewhere³⁶ in conjunction with the comparison of results of measurements on MgO and SrTiO₃ substrates. The literature values for the heat capacity are used^{31,33} to satisfy the conservation of energy for thermal flow across the interface.⁴³ The substrate is modeled as having such a large thickness (3000 nm) that the amount of heat reaching the substrate–vacuum surface is negligible on the experimental time scale. Since the grating amplitude (i.e., the amount of heat at the substrate surface) is negligible, there is no grating diffraction from this surface. In addition, the probe light field is strongly attenuated as it propagates across the film, causing negligible reflected diffraction from the back or free vacuum surface of the film. Hence only interfacial diffraction from the film–substrate interface will be considered, and the equations derived in Section 2 are entirely applicable to this situation.

C. Results and Discussion

The experimental results and model calculations shown in Fig. 5 correspond to diffraction in the forward-scattered (transmitted) and backscattered (reflected) directions, respectively. The solid lines through the data present the model thermal transient grating calculations discussed in Subsection 3.B. The oscillatory component of the signals results from the acoustic waveguide modes that are coher-

ently driven by the impulsive deposition of heat.^{38,39} The characteristics of these acoustic waves in YBCO films was discussed in detail previously.⁷ The remaining smoothly decaying portion of the signal arises from thermal redistribution of the deposited heat in the sample.

In the transmitted geometry, the signal reflects primarily the sum of the average temperature in the film and in the substrate. The majority (~90%) of the signal arises from the bulk terms (**A** and **B**) in relation (2.43). At zero temporal delay between the excitation and the probe beams, heat is entirely localized in the film near the interface because of optical absorption. Heat then rapidly diffuses into the substrate and across the film. The film has a relatively large imaginary component in the index of refraction that provides a strong resonant signal from the YBCO. The substrate, however, has no such contribution. Therefore, the signal decay arises primarily from thermal flow out of the film and into the substrate.

A significantly different picture emerges for the reflected geometry. The thermal signal has two major components. The first component arises from the interfacial terms in relation (2.42) (**E**, **H**, and **F**). The second component, which arises from bulk diffraction in the substrate, comes from terms **C** and **D** in relation (2.42). After the initial localization of heat near the film interface, the heat rapidly spreads out into the film and substrate. This causes the interfacial terms, which have contributions from both the film and substrate, to become a rapidly decaying component of the signal, characterized by a ~2-ns decay. A slowly decaying component of the signal leads to the signal baseline offset seen at long delay times (>10 ns) in the reflected signal. Roughly speaking, the offset indicates the interfacial temperature. The offset arises from the bulk diffraction terms **C** and **D** in relation (2.42). This component has a much smaller amplitude than the faster-decaying interfacial component, for two reasons. First, the bulk reflected signal has contributions only from the substrate side of the interface, so any heat that is left in the film will not contribute to the signal. Second, the substrate signal component does not have any resonantly enhanced terms resulting from absorption, in contrast to the film signal component.

The good agreement between the calculations and the data presented in this section, with a single set of parameters for both calculations, shows that the interfacial transient grating method can be used to determine the thermal diffusivity constants of two materials near an interfacial region perpendicular to the plane of the interface. Information about the nature of the microscopic thermal contact between the two materials can also be deduced. Furthermore, if one reduces the grating fringe spacing by an order of magnitude, it is possible to measure the thermal diffusivity in the plane of the interface as well.⁷

4. CONCLUSION

A detailed theoretical treatment of transient grating experiments in both the reflected and transmitted diffraction geometry from a single interface has been developed. The theory has been used to model transient grating thermal signals from a metal-film-insulating-substrate system. The high quality of the correspondence between the example experimental data and the model calculations il-

lustrates how the interfacial transient grating technique can be applied to produce detailed information on transient thermal flow near an interfacial region. This technique is not, however, limited to thermal transient grating measurements in solids. Holographic gratings experiments can be performed on virtually any gas, liquid, or solid sample interface if one chooses the appropriate grating excitation and probe wavelengths. For example, interfacial transient grating experiments performed at large and small fringe spacings on semiconductor multiple quantum wells or other layered electronic structures could lead to information on the dynamics of carriers both perpendicular and parallel to the interfacial planes between materials. The applicability of the transient grating technique to measure a variety of different physical properties suggests that the interface selective theory developed in this paper could contribute to the quantitative study of many systems.

APPENDIX A

The authors take this opportunity to correct the following typographical errors contained in a previous paper²⁸:

Equation (13a) of Ref. 28 should read as

$$u_1(x) = a_R \exp(-ik_x R x) \quad (x < 0).$$

Equation (21) of Ref. 28 should read as

$$\begin{aligned} \eta^T = & \frac{\omega^2 \cos^2 \theta \exp(-\Omega l)}{4c^2[(\epsilon - \sin^2 \theta)^{1/2}[\cos \theta + (\epsilon - \sin^2 \theta)^{1/2}]]^2} \\ & \times \left| \frac{\cos \theta - (\epsilon - \sin^2 \theta)^{1/2}}{\cos \theta + (\epsilon - \sin^2 \theta)^{1/2}} \right. \\ & \times \int_0^t \exp\left[2i \frac{\omega}{c} (\epsilon - \sin^2 \theta)^{1/2} x'\right] \Delta\epsilon(x') dx' \\ & \left. - \int_0^t \Delta\epsilon(x') dx' \right|^2. \end{aligned}$$

Equation (22) of Ref. 28 should read as

$$\begin{aligned} \eta^T = & \frac{\cos^2 \theta \exp(-\Omega l)}{4[(\epsilon - \sin^2 \theta)^{1/2}[\cos \theta + (\epsilon - \sin^2 \theta)^{1/2}]]^2} \\ & \times \left| \frac{\cos \theta - (\epsilon - \sin^2 \theta)^{1/2}}{\cos \theta + (\epsilon - \sin^2 \theta)^{1/2}} \frac{\Delta\epsilon_{(x=0)}}{2i(\epsilon - \sin^2 \theta)^{1/2}} \right. \\ & \left. - \frac{\omega}{c} \int_0^t \Delta\epsilon(x') dx' \right|^2. \end{aligned}$$

ACKNOWLEDGMENTS

We thank T. H. Gaballe and C. B. Eom for many useful discussions and for providing the high-optical-quality samples required for the experimental work. This work was supported by the National Science Foundation, Division of Materials Research (DMR90-22675), and by the U.S. Office of Naval Research, Physics Division (N00014-89-J1119). I. M. Fishman acknowledges support by the Medical Free Electron Laser Program, U.S. Office of Naval Research (N00014-91-C0170).

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