Absolute phase measurement in heterodyne detection of transient gratings

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We present a method of measuring the absolute phase in heterodyne-detected transient grating experiments. The method permits direct and sensitive characterization of the amplitude and phase of the grating parameters. We also present a convenient implementation of this technique and demonstrate its efficacy in a cuprate superconductor. © 2004 Optical Society of America

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The transient grating technique has been used successfully in a wide variety of applications, including exciton diffusion, dynamics of biomolecules, propagation of ultrasound, and thermal diffusion.[1] In this technique a pair of laser beams is interfered on a sample, producing a sinusoidally varying pattern of intensity, and hence photoexcitation density. If the presence of photoexcitations affects the index of refraction of the material, the interfering pump beams create a diffraction grating at the sample surface. The time evolution of this transient grating provides information about the relaxation and propagation of excitations created by the laser beams.

To probe the time evolution of the grating, a third beam is sent onto the sample. The probe is specularly reflected (or transmitted in the case of an optically thin sample) and diffracted from the transient grating. The intensity of the diffracted beam depends quadratically on the amplitude of the index change, δn. Because δn is often very small, the diffracted signal can be difficult to measure with standard methods of intensity (or homodyne) detection. Moreover, since photodetection of intensity destroys information about the phase, it is not possible to measure the real and imaginary parts of δn. This is a significant limitation, because information about the complex phase angle of δn is often useful in constructing a physical model of the photoexcitation process.

Heterodyne detection is an alternative detection method that enhances sensitivity and preserves the phase information contained in the diffracted probe beam.[2,3] In this technique the probe is mixed with another beam from the same laser that acts as a local oscillator (LO). One realization of the beam geometry required for coherent heterodyne detection is shown in Fig. 1. Four parallel beams, pump 1, pump 2, probe, and LO, are arranged so that the laser spots form a rectangle in a plane perpendicular to their path. When this boxcar configuration of beams is focused onto the sample under study, the difference in wave vector between the two pump beams, $\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$, is the same as the difference in wave vector between the probe beam and the LO. This geometry ensures that the diffracted probe beam emerges collinear with the specular or the transmitted LO.

In a typical experiment the beam containing both the diffracted probe and the LO is directed to a standard photodetector. The output current of the detector (I_D) is related to the total intensity:

$$I_D \propto |E_{\mathrm{LO}}|^2 + |E_S|^2 + 2\,\mathrm{Re}(E_{\mathrm{LO}}E_S{}^*)\,, \qquad (1)$$

where E_{LO} and E_S are the electric fields of the LO and diffracted probe, respectively. The last term on the right-hand side of expression (1) corresponds to the mixing of the LO and the signal-carrying beams.

We can see the improvement in the signal amplitude by analyzing the terms on the right-hand side in more detail. The first term is independent of δn. The second term is the homodyne signal, which is second order in δn. The third term is the heterodyne signal and is linearly proportional to δn. In many experiments $\delta n \approx 10^{-4}$–10^{-5}, and the third term is 4–5 orders of magnitude larger than the second term. In this range of δn, heterodyne detection can provide 4–5 orders of magnitude of improvement in the signal compared with conventional homodyne detection.

The mixing term in expression (1) can be expressed in the form $2E_{\mathrm{LO}}E_S \cos\phi$, where ϕ is the phase of the signal relative to the LO. We can vary this phase by changing the relative optical length of the probe

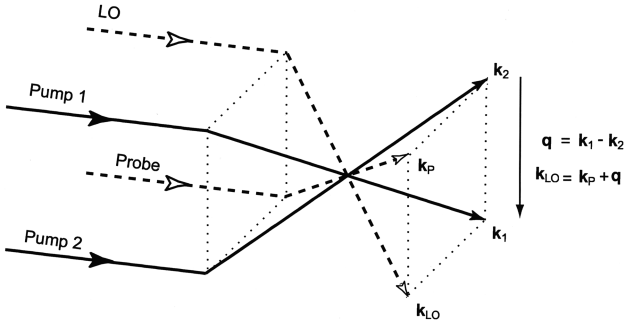

Fig. 1. Boxcar phase-matching geometry of the transient grating experiment. The pump beams that create the grating are incident along $\mathbf{k}_1$ and $\mathbf{k}_2$, and the probe and LO beams are incident along $\mathbf{k}_{\mathrm{P}}$ and $\mathbf{k}_{\mathrm{LO}}$. Since $\mathbf{k}_{\mathrm{LO}} = \mathbf{k}_{\mathrm{P}} + \mathbf{q}$, the diffracted probe beam emerges collinear with the specular or the transmitted LO.

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and LO beams. In practice, a convenient method to achieve a continuous scan of ϕ is to pass one of the beams through a thin glass coverslip placed on a rotating mount. Measuring the detector current as a function of the coverslip angle yields a direct measurement of ϕ for any position of the coverslip.

A problem encountered in transient grating experiments is that it is difficult to determine the complex phase angle of δn from a measurement of ϕ. As will be shown below, ϕ can be written as $\phi = \phi_S + \phi_{12} - \phi_{\mathrm{LP}}$,[4] where ϕ_{12} is the relative phase of the two pumps, ϕ_{LP} is the relative phase of the LO and the probe before they interact with the sample, and ϕ_S is the phase shift caused by diffraction from the transient grating. The last quantity is directly related to the phase of δn. It is clear from the expression for ϕ above that determining ϕ_S requires knowledge of $\phi_{12} - \phi_{\mathrm{LP}}$, which is difficult to measure.

Methods do exist that attempt to achieve this phase calibration. One approach is to use a material with a well-calibrated photoresponse as a reference. The most convenient example is when the sample itself has a large and well-understood index change as a result of photoinduced heating. If a calibrated thermally induced index change is not available, a different reference material in which the phase of δn is known[4,5] can be used. In addition to methods based on reference samples, direct imaging of the grating pattern by a CCD camera is possible to obtain the phase difference $\phi_{12} - \phi_{\mathrm{LP}}$.

The methods of phase calibration described above have limitations in terms of practical use. First, as mentioned above, the thermal signal is not always large enough or sufficiently well calibrated to serve as a reference. If a reference sample is used instead, any uncertainty in the phase of its photoinduced index change will introduce a systematic error in all the measurements based on this calibration. Of great importance for studies of single crystals is that the reference sample method is extremely inconvenient for low-temperature measurements performed in a reflection geometry, as the reference must be reversibly introduced into the beam with the same position and orientation as the sample. Finally, direct imaging via a CCD also requires removing the sample and performing a separate experiment to set the phase. This approach suffers from the same limitations as the techniques mentioned above, which are severe because calibration must be performed frequently to compensate for drifts resulting from temperature variation and other environmental effects.

In this Letter we introduce a new method that allows for more convenient and accurate phase calibration. It eliminates the need for a reference sample and can be used both in reflection and transmission geometries. Our method is based on the symmetry between the LO and the probe beams.[6] In the phase-matched boxcar geometry, the probe beam and the LO are interchangeable. Each beam is diffracted into the specular beam of the other. Since the two beams are symmetrical in this sense, we will refer to them as P1 and P2, rather than as probe and LO. As we show below, comparing the photodetector current as a function of ϕ (or coverslip angle) when P1 is the LO with that obtained when P2 is the LO provides the extra information required for absolute phase calibration.

The fields of the pump beams that create the grating can be written as the real parts of the complex fields $E_1 = E \exp[i(\mathbf{k}_1 \cdot \mathbf{r} + \phi_1)]$ and $E_2 = E \exp[i(\mathbf{k}_2 \cdot \mathbf{r} + \phi_2)]$. The intensity at the sample surface produced by the superposition of these fields is proportional to $|E|^2[1 + \cos(qx + \phi_{12})]$, where $\mathbf{q} = q\hat{\mathbf{x}}$ and $\phi_{12} = \phi_1 - \phi_2$. To focus on the absolute phase calibration, we treat the limit where the grating thickness is much smaller than the wavelength. In this case the grating must be probed in a reflection geometry. The extension to the thick grating case and the transmission geometry is straightforward. Assuming that the photoinduced change in reflection coefficient (δr) is proportional to the local light intensity, the reflection coefficient at the surface immediately after photoexcitation is given by $r = r_0 + \delta r[1 + \cos(qx + \phi_{12})]$, where r_0 is the equilibrium reflectivity. For times after photoexcitation the reflectivity can be written in the form $r(t) = r_0 + \delta r(t)[1 + \epsilon(t)\cos(qx + \phi_{12})]$, where ϵ is the ratio of the grating amplitude to its mean value.

As stated above, each probe beam is both diffracted and specularly reflected from the reflectivity grating. To discuss the amplitude and the phase of these waves, we rewrite the reflectivity in the form $r = r_0 + \delta r + (\delta r/2)(\eta + \eta^*)$, where $\eta \equiv \epsilon \exp[i(qx + \phi_{12})]$. The term $r_0 + \delta r$ is the coefficient of specular reflection, whereas $\delta r\eta$ and $\delta r\eta^*$ are proportional to the amplitudes for diffraction into the ± 1 diffraction orders. If we first consider probe P1, the two diffracted orders have wave vectors $\mathbf{k}_{\mathrm{P1}} - \mathbf{q}$ and $\mathbf{k}_{\mathrm{P1}} + \mathbf{q}$. Because the phase-matched geometry ensures that $\mathbf{k}_{\mathrm{P2}} = \mathbf{k}_{\mathrm{P1}} - \mathbf{q}$, the component of P1 whose wave vector is shifted by $-\mathbf{q}$ emerges collinear with the reflected wave from P2. This wave acquires phase factor η^* as it diffracts from the transient grating. Turning next to probe P2, the diffracted wave whose wave vector is shifted by $+\mathbf{q}$ is diffracted into the path of P1. This wave acquires conjugate phase factor η.

The absolute phase calibration is obtained by comparison of the intensity of the beams that emerge from the sample along the directions defined by the specular reflections of P1 and P2. To see how this comes about, we write the amplitude of the incident probe beams in phasor form $E_{\mathrm{P1,2}} = E_P \exp(i\phi_{\mathrm{P1,2}})$ (as the two beams propagate collinearly after interacting with the sample, we can ignore the spatial dependence of the phase). The amplitude in the specular path of P1 is $E_P[(r_0 + \delta_r)\exp(i\phi_{\mathrm{P1}}) + (\delta r\eta/2)\exp(i\phi_{\mathrm{P2}})]$ and the amplitude in the specular path of P2 is $E_P[(r_0 + \delta_r)\exp(i\phi_{\mathrm{P2}}) + (\delta r\eta^*/2)\exp(i\phi_{\mathrm{P1}})]$. The intensity, and therefore the photodetector current, for each of the two paths is proportional to the square of the phasor amplitude.

Neglecting terms that are second order in δr, we obtain $I_{\mathrm{P1,P2}} = I_P{}^{\mathrm{eq}} + \Delta I_{\mathrm{P1,P2}}$, where $I_P{}^{\mathrm{eq}} \propto |r_0|^2|E_P|^2$, and

$$\frac{\Delta I_{\mathrm{P1}}}{I_P{}^{\mathrm{eq}}} = \left|\frac{\delta r}{r_0}\right|[\cos(\phi_S) + (\epsilon/2)\cos(\psi - \phi_S)], \qquad (2)$$

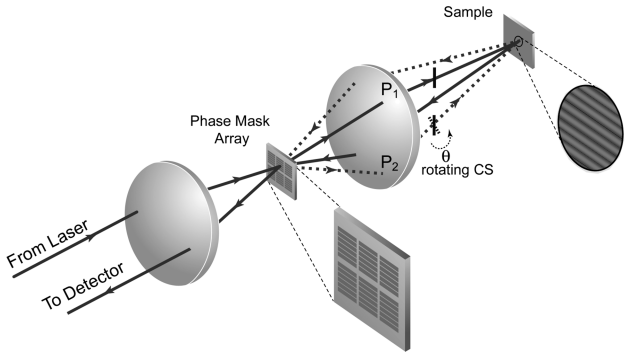

Fig. 2. Experimental setup for complete characterization of a transient grating in reflection geometry. CS, coverslip.

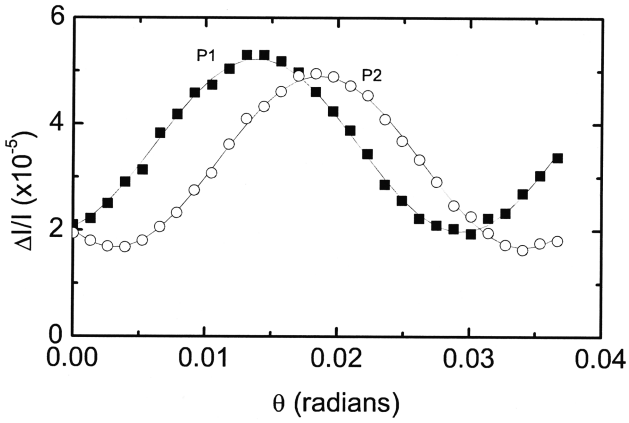

Fig. 3. Normalized photoinduced change in intensity as a function of the coverslip angle (θ) in $Bi_2Sr_2CaCu_2O_{8+\delta}$. The solid lines are fits to Eqs. (2) and (3) with $\psi(\theta) = \psi^0 + k\theta$, where ψ^0 and k are constants. The phase difference between the two cosines gives $2\phi_S = -0.96$ rad.

$$\frac{\Delta I_{\mathrm{P2}}}{I_P{}^{\mathrm{eq}}} = \left|\frac{\delta r}{r_0}\right| [\cos(\phi_S) + (\epsilon/2)\cos(\psi + \phi_S)]. \qquad (3)$$

In Eqs. (2) and (3), ϕ_S is the phase of δr with respect to r_0 and $\psi \equiv (\phi_{\mathrm{P1}} - \phi_{\mathrm{P2}}) - \phi_{12}$. Measuring ΔI_{P1} and ΔI_{P2} as a function of phase angle ψ, together with $I_P{}^{\mathrm{eq}}$, provides a complete characterization of the grating parameters, that is, $|\delta r/r_0|$, ϵ, and ϕ_S.

In Fig. 2 we show an experimental setup capable of complete transient grating characterization in a reflection geometry. Both the pump and the probe beams are split by use of a diffraction grating in the transmission mode[4,5] (for clarity, only the probe beams are shown). A spherical mirror and a plane folding mirror (represented schematically by the second lens in the figure) focus the beams onto the sample. A rotating coverslip is used to change the phase difference, $\phi_{\mathrm{P1}} - \phi_{\mathrm{P2}}$, between the two probe beams continuously. After reflection and diffraction from the surface of the sample, the probe beams pass through the same transmission grating in the reverse direction. They emerge from the grating with their collinearity restored and are sent to a photodetector.

The use of a diffractive optic beam splitter makes it possible to measure the two currents, I_{P1} and I_{P2}, without realignment of the setup. In performing the absolute phase calibration, we first block one of the reflected probes before it returns to the grating and allow the other reflected probe to reach the detector. If P2 is blocked, for example, the detector current corresponds to I_{P1}. Measurement of I_{P2} requires only that we switch the beam stop to block the reflected P1. By rotating one of the two coverslips placed in the probe beam paths we can continuously change the phase ψ. If the angle of incidence of the probe on the coverslip is close to 45°, the change in ψ introduced by a small rotation is a linear function of rotation angle θ.

We have demonstrated this technique in measurements of high-temperature superconductors.[7] According to Eqs. (2) and (3), $\Delta I_{\mathrm{P1}}(\psi)$ and $\Delta I_{\mathrm{P2}}(\psi)$ will be sinusoidal with a phase difference $\Delta\psi = 2\phi_S$. Figure 3 shows $\Delta I_{\mathrm{P1}}/I_{\mathrm{P1}}{}^{\mathrm{eq}}$ and $\Delta I_{\mathrm{P2}}/I_{\mathrm{P2}}{}^{\mathrm{eq}}$ obtained from a sample of $Bi_2Sr_2CaCu_2O_{8+\delta}$ at temperature $T = 5$ K, measured near zero time delay between the pump and the probe. The phase shift between the two detector currents is clear and yields $\phi_S = -0.48$ rad. Measuring the exact phase will help to identify the physical process causing this reflectivity change.

In summary, we have presented a new method of measuring the absolute phase of photoinduced change in reflectivity in heterodyne-detected transient grating experiments. The phase is obtained by comparison of the intensity of the two probe beams as a function of the phase change introduced by a rotating coverslip. This method resolves the long-standing problem of calibration of the phase in transient grating experiments.

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References

1. H. J. Eichler, P. Gunter, and D. W. Pohl, *Laser-Induced Dynamic Gratings* (Springer-Verlag, Berlin, 1986).
2. P. Vohringer and N. F. Scherer, J. Phys. Chem. **99,** 2684 (1995).
3. Y. J. Chang, P. Cong, and J. D. Simon, J. Phys. Chem. **99,** 7857 (1995).
4. G. D. Goodno, G. Dadusc, and R. J. D. Miller, J. Opt. Soc. Am. B **15,** 1791 (1998).
5. A. A. Maznev, K. A. Nelson, and T. A. Rogers, Opt. Lett. **23,** 1319 (1998).
6. J. P. Ogilvie, M. Plazanet, G. Dadusc, and R. J. D. Miller, J. Phys. Chem. B **106,** 10460 (2002).
7. N. Gedik, J. Orenstein, R. Liang, D. A. Bonn, and W. N. Hardy, Science **300,** 1410 (2003).