Transient anisotropy in the electron diffraction of femtosecond laser-excited bismuth

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Abstract. Laser excitation of thin bismuth films leads to a reduction in the diffraction intensity, which exhibits a characteristic angular anisotropy. The anisotropy depends on the polarization of the laser pulse and persists for approximately 150 ps. The effect clearly indicates coherent atomic motion in a preferential direction that we tentatively attribute to a transient shear deformation due to the photoelastic stress induced by the laser pulse.
1. Introduction

Over the last few years, time-resolved electron diffraction has become a valuable tool for the direct observation of femtosecond structural dynamics. Just like ultrafast x-ray diffraction, the electron diffraction counterpart combines femtosecond time resolution and atomic spatial resolution, enabling the dynamics of atomic motions to be studied directly. Examples of recent applications include ultrafast laser-induced structural phase transitions [1], cooperative rearrangement of crystalline structures [2] and dynamics of charge density waves [3].

A number of recent papers on structural dynamics have dealt with acoustic phenomena and lattice dynamics. For example, sub-picosecond acoustic transients have been observed in thin films [4–6] and in superlattice structures [7]. In these cases, femtosecond laser excitation gives rise to a rapid increase of the thermal or electronic pressure, which triggers compression and rarefaction waves. The resulting changes in the lattice constants lead to shifts of the Bragg peaks or changes in the rocking curves. Rapid laser heating also causes a short-lived decrease in the integrated reflectivity of the individual Bragg peaks. This transient Debye–Waller effect reveals the rise of the lattice temperature and enables electron–phonon energy transfer times to be determined [8].

In this paper, we report on the observation of acoustic transients not triggered by an initial change in pressure but by a higher-order direct coupling of the laser pulses with the lattice. Our electron diffraction experiments on thin bismuth films have revealed a short-lived anisotropy in the intensity of the diffraction pattern, which we attribute to a laser-driven shear deformation of the lattice. To detect this effect, a meticulous comparison of the diffraction intensity in different orders is required, because the anisotropy is rather weak. In our experiments, we take advantage of the fact that in electron diffraction several diffraction orders can be monitored simultaneously, enabling such weak changes to be established.

2. Structure of bismuth films

We summarize some structural characteristics of bismuth and of our bismuth films. Bismuth is a semimetal with $\alpha$-arsenic or A7 structure and two atoms in the primitive (rhombohedral) unit.
The lattice can be derived from the face-centred-cubic lattice by a weak rhombohedral distortion of the cubic unit cell. Figure 1 depicts a section of the reciprocal lattice relevant to the discussion of our electron diffraction experiments. It shows the (111)-plane, i.e. the plane through the origin normal to the trigonal axis ($C_3$). The dashed green arrows represent the projections of the fundamental reciprocal lattice vectors $A_1$, $A_2$ and $A_3$ onto the (111)-plane. The binary axis and the bisectrix axis are, respectively, perpendicular and parallel to the projection of $A_1$.

For an electron beam incident along $C_3$ the Bragg condition can be approximately satisfied for reciprocal lattice vectors from the (111)-plane. Accordingly, the lowest diffraction orders correspond to Miller indices $(h k l)$ given by the permutations of $(1 0 -1)$.

Epitaxially grown bismuth films on (001)-oriented substrates often exhibit a characteristic microstructure, which consists of micron-sized 90° rotated and twinned crystallites [10, 11]. Accordingly, the actual diffraction pattern from bismuth films is a superposition of the diffraction intensity from the different crystallites (domains) and is therefore expected to exhibit a twelvefold symmetry.

3. Experiment

Free-standing films of 15 nm thickness were produced by growing bismuth on a sodium chloride substrate, lifting the material off by dissolving the substrate in water, and finally depositing the films on a metallic support grid [11] with $100 \mu m \times 100 \mu m$ square openings. Laser pulses of approximately 50 fs duration at 800 nm (spectral width 28 nm) from a 1 kHz chirped pulse amplified titanium sapphire laser were used for the excitation of the films. The laser pulses were characterized using second harmonic frequency-resolved optical gating (FROG). The laser beam was incident on the back of sample at an angle of 40° with respect to the surface normal.
(trigonal axis, (111) direction) and had a spot size of approximately 1 mm. We used a pair of thin-film polarizers in conjunction with a zero-order half-wave plate to define and control the polarization of the incident laser pulse.

For time-resolved electron diffraction, we applied a pump–probe scheme in which the laser pulses excite the material and suitably delayed electron pulses probe its subsequent evolution.

The electron pulses were generated as follows. A frequency-tripled portion of the laser beam (267 nm) was used to illuminate a thin, semi-transparent silver photocathode. The photoelectrons were accelerated to 30 keV over a distance of 3 mm, passed through a 100 µm pinhole in the anode and focused to a spot size of 400 µm on a multi-channel plate (MCP) detector placed at a distance of 30 cm from the photocathode. The phosphor screen of the MCP was read out by a charge-coupled device camera.

To minimize the temporal broadening of the electron pulses during propagation, the bismuth sample was positioned only a few cm from the anode. The electron source was operated typically with a few thousands of electrons per pulse in order to reduce space charge effects. Using a channeltron detector the rms pulse-to-pulse charge fluctuation of the electron beam was measured to be 2.7%, slightly greater than the rms energy fluctuations of the third harmonic.

Based on the mean-field model proposed in [1], the electron pulse duration at the sample was estimated to be 650 fs with an energy spread of $\Delta E / E \approx 2 \times 10^{-3}$. Taking into account the angle of incidence of the laser pulse (40°), the overall temporal resolution was estimated to be 760 fs. This value is in good agreement with the experimental time resolution of better than 1 ps determined in a separate electron diffraction experiment.

The temporal overlap between the laser and the electron pulses was determined by observing the deformation of the electron beam profile produced by a laser-generated photoelectron cloud on a 200 µm metal pinhole [12]. This method enabled the delay time of the electron pulse to be controlled with an accuracy of ±250 fs.

A typical diffraction pattern from a bismuth sample is depicted in figure 2. The observed twelvefold symmetry is an indication of the microstructure of the bismuth film mentioned in section 2. One class of crystallites produces a pattern with sixfold rotational symmetry, whereas the diffraction from the other class gives a similar but 90° rotated pattern. The brightness of the diffraction spots in figure 2 is somewhat inhomogeneous because the electron beam is incident on the sample slightly off-axis.

4. Results

We will now describe the results of pump–probe experiments in which the sample was excited by the laser pulse and the diffraction intensity of the individual (1 0 −1)-type spots measured as a function of the delay time of the electron pulse. Generally, we always observed a decrease in diffraction signal, but the amount varied in a characteristic manner over the 12 diffraction spots. As an example, the red and green curves in figure 3 show the observed time dependence of the diffraction intensity in the spot circled in figure 2 in red and green, respectively (originating from the same crystalline domain). On the red spot (one o’clock position) the drop in the diffraction efficiency induced by the laser pump pulse amounts to about 4%, whereas on the green spot

4 The diffracted intensity is the signal integrated over the spot profile after subtraction of the background signal measured in the neighbourhood of the diffraction spot. No changes in the spot profile could be observed and the peak signal was proportional to the integrated signal.
Figure 2. Typical diffraction pattern from a bismuth film. The temporal evolution of the diffraction intensity in the spots circled in red and green is depicted in figure 3.

Figures 3. Diffraction signal as a function of the delay time of the electron probe pulse for a pump fluence of 1 mJ cm\(^{-2}\) (s-polarization). The green and red curves correspond to the diffraction spots of figure 2 circled in green and red, respectively. The full lines were calculated from individual data points using a smoothing procedure.

(three o’clock position) a decrease of only about 2% was measured. The decay time obtained from the red curve is approximately 3 ps.

Measurements on all of the 12 spots revealed an interesting anisotropy in the laser-induced change of the diffraction signals. Figure 4 is a polar diagram of the change in diffraction signal \(I_d\) for a probe pulse delay time of 10 ps. We plot \(1 - I_d(10 \text{ ps})/I_d(0)\) along the radius as a function

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of the azimuthal angle. \( I_d(0) \) is the diffraction signals at negative delay times before the arrival of the laser pulse. The red curve depicts the results when the laser pulses were polarized about 7° with respect to the binary axis in one type of crystallite (7° along the bisectrix axis in the 90° rotated crystallites). Zero degrees corresponds to the three o’clock position of the diffraction pattern in figure 2. The observed polar pattern clearly exhibits an angular anisotropy in the form of an oval shape with the long axis approximately along 60°.

Detailed experiments have revealed a distinct dependence of this anisotropy on the laser polarization. As an example, the results for a 90° rotation of the laser polarization are shown by the blue curve in figure 4. In this case, a pattern resembling a figure 8 shape was obtained (axis approximately along 150°). Changes in the excitation fluence had no effect on the shape of the polar patterns.

Measurements over an extended time range have established the transient character of the anisotropy. The red and green curves in figure 5 represent measurements over several hundreds of picoseconds for the one o’clock and the three o’clock positions, respectively. At three o’clock an approximately constant signal decrease of about 4% was measured. On the other hand, a stronger initial decrease was observed at one o’clock. This signal relaxes in about 150 ps and approaches the green curve of the three o’clock signal. The example shows that the anisotropy in the electron diffraction disappears in a few hundreds of picoseconds. After the relaxation of the anisotropy, we are left with an isotropic decrease of the diffraction intensity attributable to the Debye–Waller effect [8].

**Figure 4.** Polar diagrams plotting the change in diffraction intensity in per cent as a function of the azimuthal angle at 10 ps delay time for s- and p-polarized laser excitation (red and blue, respectively).
5. Discussion

What kind of structural change could be responsible for the observed angular anisotropy in the electron diffraction after laser excitation?

Short-pulse laser excitation usually leads to a rapid increase in the lattice temperature, which gives rise to a decrease in the diffraction signals via the Debye–Waller effect. In bismuth, the random thermal motion of the lattice atoms is isotropic and so is the Debye–Waller effect, e.g. in our case the decrease in diffraction signal attributable to the Debye–Waller effect is the same for all permutations of (1 0 −1) diffraction orders. It could be argued that for oblique incidence the fraction of the laser energy absorbed by the specimen depends on the laser polarization. But all diffraction orders would be affected likewise and no anisotropy is expected.

On the other hand, oscillations or displacements of the lattice atoms in some preferential direction would give rise to an angular anisotropy. Accordingly, our experimental observations could be accounted for by a mechanism capable of both driving some directional atomic motion and controlling the direction via the laser polarization. Clearly, thermally excited atomic motion can be ruled out and a higher order direct coupling between the light and the lattice must be considered.

In our experiments, we measure diffraction corresponding to reciprocal lattice vectors perpendicular to the trigonal axis, and only atomic displacements perpendicular to this axis can be observed. In the following, we discuss Raman- and Brillouin-type interactions as possible mechanisms for the generation of such atomic displacements. Our principal goal is to qualitatively explain the observed angular anisotropy and its dependence on laser polarization.

5.1. Raman interaction

As an example, we take the excitation of near zone centre transverse optical phonon (TO) modes by impulsive stimulated Raman scattering (ISRS). In bismuth, these TO phonon modes possess...
$E_g$ symmetry and represent a coherent motion of the two basis atoms of the unit cell in the direction perpendicular to the trigonal axis. Let $U_{E_g}$ denote the amplitude of the collective atomic displacement generated by linearly polarized laser pulses. The resulting change in the electron diffraction intensity is determined through the structure factor $S_{hkl}$ by the projection of the displacement vector $U_{E_g}$ on the reciprocal lattice vector $G_{hkl}$:

$$|S_{hkl}|^2 \propto \cos^2 \left( G_{hkl} \cdot U_{E_g} \right).$$

(1)

According to equation (1) the excitation of TO phonon modes leads to a decrease in diffraction intensity, which would be the highest for diffraction spots corresponding to $G_{hkl}$ parallel to $U_{E_g}$. In a polar diagram plotting the change in diffraction intensity as a function of the angle between $G_{hkl}$ and $U_{E_g}$, a figure-8-shaped pattern directed along $U_{E_g}$ would be expected. The direction of $U_{E_g}$, on the other hand, is linked with the polarization of the laser pulses via the Raman tensor.

While the Raman model predicts anisotropy and a dependence on the laser polarization, it fails to explain the observed time dependence. Firstly, the time to build up the TO-mode is given by the laser pulse duration, which is much shorter than our experimental time resolution. The latter is determined by the duration of the electron probe pulses. Thus, for a Raman process the fall time of the diffraction signal is expected to be less than a picosecond rather than several picoseconds as observed. Secondly, and more importantly, the measured relaxation time of the anisotropy is approximately 150 ps, whereas the reported lifetime of the TO-mode is less than 1 ps [13]. One could argue that the electron diffraction experiments primarily measure the lifetime of the near zone centre population of the TO phonon branch. The lifetime $\tau'$ of the population could be greater than the lifetime $\tau$ of the individual modes [14]. However, the TO phonon relaxation is believed to be dominated by three-phonon-interaction processes whereby the TO-phonon decays into two acoustic phonons [15]. Under these circumstances, we expect $\tau' \approx \tau$. Thus, the observed time dependence of the anisotropy argues against the Raman model.

5.2. Brillouin interaction

Another way of creating a coherent directional motion is the Brillouin mechanism, i.e. photoelastic (or electrostrictive) interaction of light with acoustic modes. We limit the discussion to transverse acoustic modes propagating in the $C_3$ direction. These modes represent shear deformations of the lattice with atomic displacements perpendicular to $C_3$, i.e. the type of displacement that would produce the observed anisotropy in the electron diffraction.

The photoelastic interaction of light with acoustic modes can be generally described by the equation [16]

$$\rho \frac{\partial^2}{\partial t^2} U_i - c_{ijkl} \frac{\partial^2}{\partial x_j \partial x_k} U_i = \frac{n^4}{8\pi} p_{ijkl} \frac{\partial}{\partial x_j} (E_k E_l).$$

(2)

The $U_i$ are the components of the displacement, $\rho$ is the mass density and $p_{ijkl}$ and $c_{ijkl}$ are the elements of the fourth rank photoelastic tensor and elastic stiffness tensor, respectively. $E_k$ and $E_l$ are the components of the electric field of the laser pulse and $n$ is the refractive index.

When equation (2) is transformed to a coordinate system with the $x_1$-, $x_2$- and $x_3$-axes along the binary, bisectrix and trigonal axes, respectively, we obtain

$$\frac{\partial^2}{\partial t^2} U_{TA1} - v_{TA1}^2 \frac{\partial^2}{\partial x_3^2} U_{TA1} = F_{TA1}. $$

(3)
Equation (3) describes TA modes propagating along \(C_3\) (\(x_3\)-axis) with displacements \(U_{TA1}\) along the \(x_1\)-axis (binary, see figure 1). \(v_{TA}\) is the transverse sound velocity along \(C_3\). The driving term \(F_{TA1}\) is a function of the photoelastic coefficients and of the three angles \(\theta, \varphi, \psi\): the incidence angle, the angle between the plane of incidence and the binary axis, and the angle between the electric field vector and the normal of the plane of incidence. For a linearly polarized incident wave \(\psi = 0\) and \(\psi = \pi/2\) correspond to s-polarization and p-polarization, respectively.

We have carried out a detailed theoretical study of the angular behaviour of the photoelastic driving forces. For the general case of oblique incidence, the driving force \(F_{TA1}\) is too complicated to be presented here in full length. However, for normal incidence, equation (3) is greatly simplified:

\[
\frac{\partial^2}{\partial t^2} U_{TA1} - v_{TA}^2 \frac{\partial^2}{\partial x_3^2} U_{TA1} = \frac{n^4}{8\pi \rho} p_{\text{eff}} \sin 2\varphi \frac{\partial}{\partial x_3} E_0^2(x_3,t). \tag{4}
\]

Here, \(p_{\text{eff}} = (P_{11} - 2P_{44} - P_{12})\) is the effective photoelastic coefficient, where \(P_{11}, P_{44}\) and \(P_{12}\) are the photoelastic coefficients in Voigt notation for cubic crystals. The small rhombohedral distortion is neglected, and the bismuth lattice is treated as face-centred cubic. Equations similar to (3) and (4) can be written down for acoustic modes polarized along the \(x_2\)-axis (bisectrix).

The driving terms \(F_{TA1}\) and \(F_{TA2}\) determine the direction of the acoustic displacements. Equation (4) shows that for normal incidence a rotation of the laser polarization by \(\varphi\) results in a rotation of the displacement by \(2\varphi\). Recall that the discussion following equation (1) has shown that a unidirectional atomic displacement gives rise to a figure-8-shaped anisotropy. Accordingly, for normal incidence the figure-8 pattern is expected to rotate twice as fast as the laser polarization. In particular, a 90° rotation of the laser polarization would not change the electron diffraction pattern, in disagreement with the experiments. However, our detailed examination of the angular dependence of the photoelastic driving forces reveals that for oblique incidence a much greater variety of shapes and rotational behaviour can occur. The actual oblique angle of incidence has to be taken into account to explain our experimental observations.

It remains to describe the behavior of the complete twelvefold diffraction patterns, i.e. to take into account the domain structure (two different classes of crystallites) of the specimen. To obtain the acoustic displacement fields for the other domain, the acoustic wave must be analysed in a reference frame rotated around \(C_3\) by 90°.

A result of such an analysis is presented in figure 6, which shows a representative pair of calculated twelvefold polar patterns illustrating the changes in diffraction signal caused by photoelastically excited TA modes. The anisotropy patterns also depend on the ratio of the photoelastic coefficients. Because the values for bismuth are not available, a combination of coefficients that produces patterns similar to the observed ones was taken. An isotropic (Debye–Waller) contribution of 30% was included in figure 6.

The example in figure 6 demonstrates that the photoelastic model is able to explain qualitatively the observed angular anisotropy in the electron diffraction on the laser polarization. In particular, the example demonstrates the effect of a 90° rotation of the laser polarization, which carries the blue pattern over to the red one.

We have shown that an analysis of the photoelastic driving forces is sufficient to explain the anisotropy effects qualitatively. For a more complete account of the changes in the electron diffraction caused by a photoelastic process, the spatial and temporal characteristics of the...
Figure 6. Calculated polar diagrams plotting the change in diffraction intensity as a function of the azimuthal angle. The laser polarizations in the red and blue patterns are perpendicular.

generated displacement field must be known. As before, we restrict ourselves to transverse acoustic modes propagating in the $C_3$ direction. For normal incidence, we are left with equation (4), a one-dimensional inhomogeneous wave whose solutions are well known [17]. It is shown in the appendix that with some additional simplifying assumptions the solutions take on a very simple form. The displacement waves can be represented by Heaviside $\Theta$-functions:

$$U_{TA1}(x,t) = U_0 \sin 2\phi \Theta(v_{TA}t - x),$$  

$$U_{TA2}(x,t) = U_0 \cos 2\phi \Theta(v_{TA}t - x).$$

In the layer $x < v_{TA}t$ the material is uniformly displaced by an amount $U_0$ in the direction perpendicular to the trigonal axis. Material with $x > v_{TA}t$ remains untouched. Note that the direction of the displacement varies with the laser polarization: $\phi$ is the angle between the electric field of the laser and the binary axis. Laser polarization corresponding to $\phi = \pi/4$ drives a motion along the binary axis, whereas $\phi = 0$ or $\pi/2$ produces a displacement in the direction of the bisectrix.

To discuss the changes in the electron diffraction intensity produced by the excitation of TA modes, we refer to figure 7 that depicts a snapshot of the displacement wave $U_{TA}$ predicted by the Brillouin model. The laser pulse strikes from the left and $L$ is the thickness of the material layer. Using kinetic diffraction theory, the total scattered amplitude is a coherent superposition of the contributions from the two sublayers A and B. The scattering conditions for A and B are identical except for a phase shift $\delta$ for the scattering contribution from A. The phase shift
is given by $\delta = U_0 \mathbf{G}_{hkl} \cdot \mathbf{e}$, where $\mathbf{e}$ is the unit vector in the direction of the displacement. The structure factor can be written as

$$\frac{|S(\mathbf{G}_{hkl})|^2}{|S_0(\mathbf{G}_{hkl})|^2} = \frac{a^2 + b^2 + 2ab \cos \delta}{(a + b)^2}, \quad a = v_{TA} t, \quad b = L - v_{TA} t,$$

where $S_0$ is the structure factor without excitation.

No change in the diffraction intensity occurs for diffraction orders corresponding to reciprocal lattice vectors $\mathbf{G}_{hkl}$ perpendicular to $\mathbf{e}$. Otherwise we have a decrease in the diffraction intensity for all orders. In the present example, only the wave starting from the front surface is considered. In this case, the minimum of the diffraction intensity occurs when the displacement wave reaches the centre of the layer.

Finally, we discuss the temporal evolution of the diffraction intensity. In our simple model, the acoustic wave starts from the surface and travels back and forth in the layer after being reflected from the back at time $t = L/v_{TA}$. The sequence of reflections would lead to a corresponding periodicity in the diffraction intensity. The transverse acoustic sound velocity in bismuth in the $C_3$ direction is $v_{TA} \approx (C_{11} - C_{12} + C_{44})/3\rho = 1310 \text{ m s}^{-1}$. The $C_{ik}$ are the elastic stiffness constants and $\rho$ is the mass density. Taking the elastic constants from [18] the transit time is calculated to be $T = L/v_{TA} = 11 \text{ ps}$ ($L = 15 \text{ nm}$). However, the experimental error of the diffraction data was too large and prevented a verification of the predicted oscillations.

The purpose of this discussion was to provide qualitative arguments in support of the Brillouin mechanism. A detailed quantitative explanation of the observed time evolution and of the detailed dependence of the anisotropy on the angle of incidence and the polarization of the laser pulse would be beyond the scope of this paper.

For a more realistic modelling of the temporal evolution, one should also take into account that in the absence of absorption we expect waves both starting from the front and the back of the film. Furthermore, it can be seen from (A.3) that in the presence of absorption we have an additional driving term and therefore the displacement wave is no longer a simple step function.
6. Conclusions

Femtosecond laser excitation produces a transient angular anisotropy in the electron diffraction from thin bismuth films. The principal features of this phenomenon are: (i) laser excitation gives rise to a decrease in the diffraction intensity; (ii) the decrease varies with the azimuthal angle of the scattering vector; (iii) the angular anisotropy in the diffraction intensity changes with the laser polarization; and (iv) the anisotropy disappears within approximately 150 ps.

The effect is interpreted as a transient coherent motion of the lattice atoms in a direction perpendicular to the trigonal axis. We have discussed Raman and Brillouin interactions as a possible mechanism for the generation of the coherent lattice motion. Both mechanisms could, in principle, explain an anisotropy in the diffraction intensity and its dependence on the laser polarization. However, a significant shortcoming of the Raman model is the failure to explain the $\approx 150$ ps lifetime of the anisotropy, because the TO phonon lifetimes in bismuth are much shorter. In contrast, the observed relaxation time of the anisotropy falls in the range of the expected acoustic phonon relaxation times and therefore supports the Brillouin model.

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Appendix

To discuss the spatial and temporal characteristics of the acoustic displacement field, we write the laser field as follows:

$$E_0^2(x,t) = A_0^2(t - x/c) e^{-\alpha x} \cos^2 [k (x - ct)],$$  \hspace{1cm} \text{(A.1)}

where $k$ is the wave vector and $\alpha$ takes into account the optical absorption of the material. The subscript on $x_3$ has been dropped ($x_3 \rightarrow x$).

The pulse intensity is $I(t - x/c) = \frac{c^n}{8\pi} A_0^2 (t - x/c)$. Because in the case of interest the transit time through the sample $L/c$ is very short, we have $I(t - x/c) \approx I(t)$. The rapidly varying terms in $\text{(A.1)}$ are neglected. Accordingly, $\cos^2$ is replaced by $1/2$.

Using the Heaviside $\Theta$-function to describe a sample occupying the half-space $x > 0$, equation $\text{(A.1)}$ can now be cast in the form

$$\frac{\partial^2}{\partial t^2} u_{\text{TA}1} - v_{\text{TA}}^2 \frac{\partial^2}{\partial x^2} u_{\text{TA}1} = \frac{n^3 p_{\text{eff}}}{2\rho c} I(t) \frac{\partial}{\partial x} \Theta(x) e^{-\alpha x}.$$  \hspace{1cm} \text{(A.2)}

When the optical absorption is neglected ($\alpha \ll 1$), the shear strain $\sigma_{\text{TA}1} = \frac{\partial u_{\text{TA}1}}{\partial x}$ corresponding to the solution of $\text{(A.2)}$ has a particularly simple form. The strain pulse assumes the shape of the laser pulse and travels with the acoustic velocity towards the bulk:

$$\sigma_{\text{TA}1}(x,t) \propto I(t - x/v_{\text{TA}}).$$  \hspace{1cm} \text{(A.3)}
Because the speed of sound is much smaller than the speed of light, the strain pulse corresponds to a very good approximation, to a delta function $\delta(t - x/v_{TA})$. Accordingly, the displacement wave can be written as

$$U_{TA1}(x, t) \propto U_0 \Theta(t - x/v_{TA}),$$

where $U_0$ is given by

$$U_0 = \frac{n^3 p_{eff} F_0}{2\rho v_{TA} c},$$

$F_0$ is the energy fluence of the laser pulse. For a crude estimate of the displacement $U_0$, we use the following parameters: refractive index $n = 2.8$, $p_{eff} = 0.1$ [19]; $F_0 = 10^7$ erg cm$^{-2}$, mass density $\rho = 9.78$ g cm$^{-3}$ and $v_{TA} = 10^5$ cm s$^{-1}$. We obtain $U_0 \approx 5 \times 10^{-10}$ cm = 0.005 nm. The phase shift for the $(1 0 -1)$-type diffraction orders is $\delta = 2\pi u_0/d_{hkl} \approx 0.05\pi$, where $d_{hkl} = 0.228$ nm. Using equation (A.5), our estimate of $U_0$ suggests a decrease in diffraction intensity by a fraction of one per cent.

References

[1] Siwick B J, Dwyer J R, Jordan R E and Miller R J D 2003 Science 302 1382–5
[2] Baum P, Yang D S and Zewail A H 2007 Science 318 788
[3] Eichberger M, Schäfer H, Krumova M, Beyer M, Demsar J, Berger H, Moriena G, Sciaiini G and Miller R J D 2010 Nature 468 799
[4] Shymonovich U, Nicoul M, Kähle S, Lu W, Tarasevitch A, Zhou P, Wietler T, Horn von Hoegen M, von der Linde D and Sokolowski-Tinten K 2010 Materials Research Society. Symp. Proc. vol 1230E MM06-03
[5] Harb M, Peng W, Sciaiini G, Hebeisen C T, Ernstorfer R, Eriksson M A, Lagally M G, Kruglik S G and Miller R J D 2009 Phys. Rev. B 79 094301
[6] Park H, Wang X, Nie S, Clinite R and Cao J 2005 Solid State Commun. 136 559–63
[7] Bargheer M, Zhavoronkov N, Grisai Y, Woo J C, Kim D S, Woerner M and Elsaesser T 2004 Science 306 1771
[8] Ligges M, Rajkovic I, Zhou P, Posth O, Hassel C, Dumpich G and von der Linde D 2009 Appl. Phys. Lett. 94 101910
[9] Hofmann P h 2006 Prog. Surf. Sci. 81 191–245
[10] Hattab H, Zubkov E, Bernhart A, Jnawali G, Bobisch C, Krenzer B, Acet M, Möller R and Horn-von Hoegen M 2008 Thin Solid Films 516 8227–31
[11] Payer T h, Rajkovic I, Ligges M, von der Linde D, Horn-von Hoegen M and Meyer zu Heringdorf F-J 2008 Appl. Phys. Lett. 93 093102
[12] Dolocan A, Hengsberger M, Neff H J, Barry M, Cirelli C, Greber T and Osterwalder J 2006 Japan. J. Appl. Phys. 45 285
[13] Ishioka K, Kitajima M and Misochko O V 2006 J. Appl. Phys. 100 093501
[14] Laubereau A, von der Linde D and Kaiser W 1972 Phys. Rev. Lett. 28 1162
[15] Cowley R A 1968 Rep. Prog. Phys. 31 123
[16] Kroll N 1965 J. Appl. Phys. 36 34
[17] Korn G A and Korn T M 1961 Mathematical Handbook for Scientist and Engineers (New York: McGraw-Hill)
[18] Lichnowski A J and Saunders G A 1976 J. Phys. C: Solid State Phys. 9 927
[19] Nye J F 1957 Physical Properties of Crystals (Oxford: Oxford University Press) p 244

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