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AN EXPERIMENTAL SETUP FOR PROBING THE THERMAL PROPERTIES OF DIAMOND REGARDING ITS USE IN AN XFELO*

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Abstract

This work presents an optical pump-probe setup for measuring the thermal evolution of diamond crystals at cryogenic temperatures under the heat load conditions of an X-ray free-electron laser oscillator (XFELO). As an XFELO is based on a cavity using diamond Bragg reflectors and these reflectors are subjected to intense heat loads during operation, the correct understanding of the thermal evolution in diamond plays a major role in the correct modeling of an XFELO. *Stoupin et al.* [1] did a room temperature x-ray diffraction measurement on the nanosecond transient thermal response of diamond to an optical pulse. The measurements presented in this paper for the first time incorporate effects due to the very short penetration depth of only a few µm of an XFELO pulse in combination with the high mean free path in diamond at cryogenic temperatures. While at room temperature the heat equation based on Fourier's law accurately fits the measured results, this vastly changes due to the onset of ballistic processes at cryogenic temperatures. These changes, which are hard to predict theoretically, show the necessity of measurements of the thermal evolution in diamond with special regard to a correct mimicking of the heat load in an XFELO.

INTRODUCTION

Current hard X-ray free-electron laser (FEL) facilities all use the self-amplified spontaneous emission (SASE) scheme for operation. While these sources produce very brilliant femtosecond X-ray pulses with excellent transverse coherence, they suffer from a lack of longitudinal coherence. A promising approach for reaching full longitudinal coherence in the hard X-ray regime proposed by *Kim et al.* in 2008 [2] is the X-ray free-electron-laser oscillator (XFELO). This scheme is based on using a rather short undulator with a length of less than 15 m and a highly reflective cavity based on very pure diamond crystals serving as Bragg reflectors. As these Bragg reflectors also act as spectral filters an XFELO promises a spectral bandwidth in the order of the crystals bandwidth ($\Delta\omega/\omega \approx 10^{-5} - 10^{-7}$) and therefore orders of magnitude better than SASE-FELs. Furthermore, as the radiation field is built up over many cavity round trips, very low shot-to-shot fluctuations can be expected, even making the XFELO a promising candidate for X-ray quantum optics (XQO) [3]. With the recently commissioned European XFEL the realization of an XFELO becomes in reach. This is due to the facility's excellent electron beam properties and especially due to its very high bunch repetition rate of 4.5 MHz in pulsed-mode [4] which enables resonator lengths of only 33 m.

A major issue one needs to address when dealing with an XFELO is the effect of the light-matter interaction between the X-ray field and the Bragg reflectors. This is due to the high requirements for the angular and spatial stability [5, 6] as well as the necessity of very stable Bragg conditions.

As shown by *Zemella et al.* in 2012 [7] even an XFELO with a designedly limited saturated pulse energy of ≈ 250 µJ circulating with a rate of 4.5 MHz leads to a considerable heating of the Bragg reflectors and thereby to their thermal expansion. This thermal expansion may lead to vibrations of the crystal, change of the wavelength satisfying the Bragg's law [7] and the generation of ultrasonic pulses [1,8]. The last effect is studied by *Bahns et al.* [9] also at this conference.

Owing to the importance of keeping these effects as low as possible, it has already been concluded that diamond at cryogenic temperatures is the ideal candidate for an XFELO [10,11], due to its very high thermal conductivity, its low thermal expansion coefficient as well as high radiation hardness and a Bragg reflectivity over 99%. Nonetheless, even with diamond at 50 K, heating of the crystals does not seem negligible [7]. Consequently, in order to properly predict the behavior of an XFELO at the European XFEL and the effect of the thermal load, the diamond reflectors' thermal response need to be understood and especially measured as will be shown in the following. Our approach is to mimic an XFELO by a UV laser which deposits roughly the same energy into a diamond crystal at the same penetration depth as an saturated XFELO pulse. Such an experimental setup is presented in this work.

QUASI-BALLISTIC HEAT TRANSPORT

As discussed above, diamond is the ideal candidate for Bragg reflectors in an XFELO. However, when treating thermally highly conductive materials, especially at low temperatures where the phonon-phonon Umklapp scattering is freezing out, one has to take into account size effects which lower the predictive power of the heat equation based on Fourier's law. The latter is based on the assumptions of local thermal equilibrium and time- and length scales of interest larger than the typical scattering time or mean free path of a phonon, respectively. As the mean free path in diamond is of the order of hundreds of microns at $T = 50$ K and still of the order of tens of microns at $T = 100$ K, these assumptions begin to fail and size dependent ballistic processes begin to occur.

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There are two different size effects to consider. The first is the so called "Gradient Effect": When a temperature gradient ∇T is varying quickly on length scales comparable to the mean free path, ballistic effects begin to have an considerable influence. Traditionally one would simulate these effect by the use of the computationally expensive Boltzmann-Transport equation or the simplified McKevley-Shockley flux method (MSM) yielding comparable results [12]. But as has been pointed out by *Maassen and Lundstrom* in 2015 [13, 14], the MSM for the heat transport can be rewritten as computationally much cheaper diffusion equations. This is under the premise one knows the physically correct boundary conditions as well as the energy dependence of the scattering parameter $\tau(\epsilon)$. These boundary equations as well as the exact $\tau(\epsilon)$ are very hard to estimate theoretically, as will be shown in the following paragraph. While in the stationary case Fourier's law

$$\boldsymbol{q} = -\lambda_{th} \nabla T,$$

with the thermal heat flux density $\boldsymbol{q}$ and the thermal conductivity λ_{th}, and the corresponding heat equation hold true [13], they begin to fail in the transient case of the gradient effect [14] for the reasons discussed above. In this case, which also describes the situation in an XFELO, Fourier's law can still be applied. But one has to keep in mind, that the thermal conductivity in the formula is physically incorrect and Fourier's law only apparently holds. Nonetheless in this work we use it, as the deviation of the derived $\lambda_{th}^{(\mathrm{app})}$ from the literature bulk values gives a good estimate of the significance of ballistic processes.

The other size effect is the boundary-scattering at the crystal surfaces, which lead to a reduction of the phonons' mean free path. Consequently Fourier's law would also yield a reduced apparent thermal conductivity. As the temperature dependent energy ϵ spectrum of the mean free path $l_{\mathrm{mfp}}(\epsilon)$ or scattering time $\tau(\epsilon)$ of the phonons can span orders of magnitude, it is hard to predict how the individual phonon modes are scattered at the surface [15, 16], making an accurate estimate of the effect hard to achieve. This effect is important for an XFELO, especially when one plans to couple out the radiation by using a very thin ($\sim 40\,\mu\mathrm{m}$) and therefore partly transmissive diamond crystal [7]. But it is of no significance at the temperatures experimentally probed in this paper.

EXPERIMENTAL SETUP

As one can see from the discussion in the previous section, it is of great importance to conduct thermal measurements when it comes to properly modeling the heat transport under conditions given for the XFELO. For this purpose an optical pump-probe experiment displayed in Fig. 1 was developed. In particular, a nanosecond 213 nm UV laser with a pulse energy of 2.5 µJ - which is about the absorbed pulse energy in a saturated XFELO [7] - is being absorbed by a diamond crystal, which causes a time dependent distortions of the temperature T inside the solid.

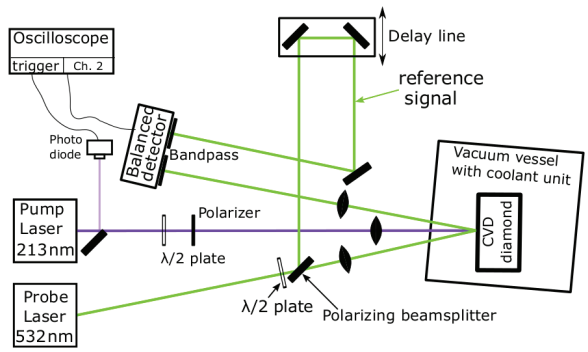

Figure 1: Scheme of the experimental setup. A nanosecond UV pump laser providing the pulse energy characteristics of an XFELO introduces a change in temperature in the sample, a 300 µm thick CVD diamond single crystal. The change in temperature is probed by the reflection of a green cw laser. A balanced photo detector amplifies the difference between the reflected signal and a reference signal. A fast oscilloscope averages the signal's time evolution over many pump pulses.

The temperature couples to the refractive index $\tilde{\eta} = \eta' + i\eta''$ and therefore to the diamond optical properties. One can directly relate a change in reflectivity [17]

$$\frac{\Delta R}{R} = \frac{4(\eta'^2 - \eta''^2 - 1)\Delta\eta' + 8\eta'\eta''\Delta\eta''}{\left[(\eta'+1)^2 + \eta''^2\right]\left[(\eta'-1)^2 + \eta''^2\right]} \approx \underbrace{\left(\frac{1}{R}\frac{\delta R}{\delta \Delta T}\right)}_{k_T} \Delta T$$

to a change in the refractive index $\Delta\tilde{\eta} = \Delta\eta' + i\Delta\eta''$ and therefore to a change in temperature ΔT. The above linear approximation is valid for temperature changes of a few degree. In this work, a green 532 nm cw-laser is used to measure the change in reflectivity and with the knowledge of $k_T \Delta T$ induced by the UV pump laser can be determined. The physical time resolution is mainly determined by the photo diode's rise time and the bandwidth of the detection system.

As the thermal gradient resulting from the pump pulse is long compared to the probe lasers' wavelength, the probe signal does not notice abrupt changes in the temperature dependent refractive index, which would be a premise for reflection. Consequently no internal reflections occur and the measured change in reflectivity is directly proportional to the temperature at the sample surface.

A challenge in this experimental setup is the rather bad signal to noise ratio. This is caused by a low thermoreflectance calibration coefficient k_T relating a big change in ΔT to a comparably low ΔR. To overcome this bad signal to noise ratio each $\Delta R(t)$ curve has to be averaged over many pump pulses.

In order to get information on the thermal evolution at cryogenic temperatures, the sample is located in a vacuum chamber attached to a helium pulse tube cooler. With this cooling unit, temperatures down to ~ 50 K can be reached.

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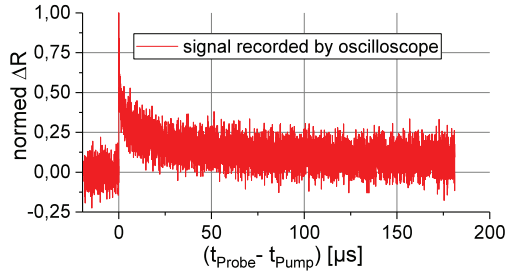

Figure 2: Time evolution of the change in reflection at T_0 = 297 K showing a slow µs to ms long contribution to the crystals recovery back to its initial state. This contribution results from excitations of long living electron states by the pump laser. The bad signal to noise ratio comes from a low number of averages.

RESULTS

Figure 2 displays an exemplary measurement for the long time evolution of the change in reflectivity after the pump pulse at T_0 = 297 K. The measurement shows a slow decline of the reflectivity on the scale of approximately 100 µs after the absorption of the pump pulse. This is much slower then the expected thermal response on the timescale of a few hundred nanoseconds. The effect can probably be traced back to the excitation of long living electronic states in the diamond after the absorption, which also changes the sample's refractive index $\tilde{\eta}$. In order for it not to disturb the analysis of the thermal response, in the following an offset is subtracted from the measurement signal. This is justified as the slow decay appears constant on the much shorter timescale of the thermal processes. Also one has to note, that these long living electronic states do not disturb the XFELO operation, as Bragg reflection is much less sensitive to the electronic structure than optical reflection.

Figure 3 displays measurements at two different temperatures: (a) T_0 = 297 K and (b) T_0 = 150 K. In addition, a plot of the derived thermal conductivities versus the temperature is shown (c). In both measurements (a) and (b) the signals are cleared from an offset as discussed above. Both measurements (a) and (b) still exhibit a noisy background, but a clear evolution of the reflectivity on the nanosecond scale is apparent. This evolution is fitted to a model derived from Fourier's law. For the room temperature case (a) and T_0 = 250 K the derived λ agrees well with the literature value (see Fig. 3(c)). This can be seen as a proof of principle measurement for the validity of the experimental setup, as one would not yet expect ballistic effects to play a role at these temperatures. However, the measurements at lower temperatures, especially at T_0 = 150 K begin to show strong deviations from the literature bulk values. This is in agreement with the discussion of the "gradient effect", as for T = 150 K the middle mean free path $l_{\mathrm{mfp}} \approx 15$ µm is of the same order as the pump pulse's penetration depth.

Another important consideration is that at room temperature a significant portion of the thermally induced shift in reflectivity remains after 220 ns, while at T_0 = 150 K,

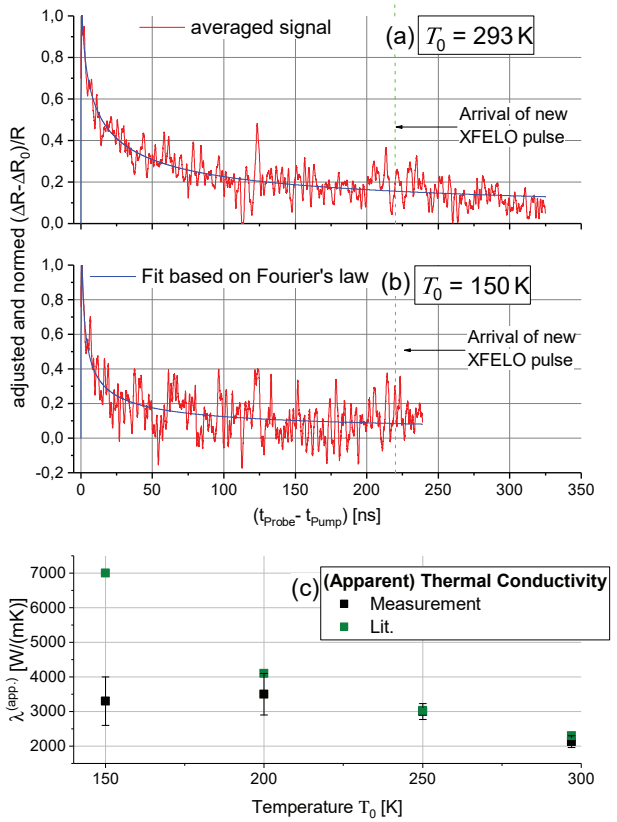

Figure 3: Nanosecond scale time evolution of the change in reflectivity for (a) T_0 = 297 K and (b) T_0 = 150 K. Following the discussion regarding Fig. 2 the results are corrected by a constant offset. Because of a shorter measurement time, (b) exhibits a noisier background. The signals are each fitted to a model based on Fourier's law. A plot of the derived thermal conductivities against the temperature (c) is displayed together with their bulk literature values [18].

ΔR has declined significantly more. This shows the usefulness of cooling for avoiding consecutive heating of the Bragg reflectors and the resulting disturbances in operation [7] in an XFELO with a repetition rate of 4.5 MHz.

CONCLUSION AND OUTLOOK

An tabletop setup for measuring the effect of thermal load on diamond Bragg crystals in an XFELO was developed. The validity of the measurement method was demonstrated by proof of principle measurements at room temperature and T_0 = 250 K, which yield thermal conductivities λ_{th} agreeing well with bulk literature values. However, with decreasing temperature the calculated $\lambda_{\mathrm{th}}^{(\mathrm{app.})}$ from Fourier's law begin to differ considerably from the respective bulk values. This can be traced back to the growing contribution of ballistic processes to the heat conduction at decreasing temperatures. This supports the necessity of experimentally measuring diamonds' thermal properties at low temperatures under XFELO conditions. Also, the measurement exhibited a not fully declined thermal excitation after 220 ns, confirming the idea of a pile up of thermal energy in a 4.5 MHz XFELO.

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In the future measurements at crystals with varying thickness and at additional temperatures need to be conducted, especially at $T_0 = 50\,\mathrm{K}$ and $T_0 = 100\,\mathrm{K}$, where additionally effects due to boundary scattering are expected. Finally, these results need to be implemented into the modeling of an entire XFELO.

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REFERENCES

[1] S. Stoupin *et al.*, "Direct observation of dynamics of thermal expansion using pump-probe high-energy-resolution x-ray diffraction," *Phys. Rev. B*, vol. 86, p. 054301, Aug 2012.

[2] K.-J. Kim, Y. Shvyd'ko, and S. Reiche, "A proposal for an x-ray free-electron laser oscillator with an energy-recovery linac," *Physical Review Letters*, vol. 100, jun 2008.

[3] B. Adams and K.-J. Kim, "X-ray comb generation from nuclear-resonance-stabilized x-ray free-electron laser oscillator for fundamental physics and precision metrology," *Physical Review Special Topics - Accelerators and Beams*, vol. 18, mar 2015.

[4] "European XFEL," located at http://www.xfel.eu/overview/facts_and_figures/.

[5] S. Stoupin *et al.*, "Nanoradian angular stabilization of x-ray optical components," *Review of Scientific Instruments*, vol. 81, no. 5, p. 055108, 2010.

[6] C. Maag, J. Zemella, and G. J. Rossbach, "Numerical studies of the influence of the electron bunch arrival time jitter on the gain process of an XFEL-oscillator for the European XFEL," in *Proc. of FEL'15*, Daejon, Korea, TUP032.

[7] J. Zemella, J. Rossbach, C. Maag, M. Tolkiehn, and H. Sinn, "Numerical simulations of an XFELO for the European XFEL driven by a spent beam," in *Proc. of FEL'12*, Nara, Japan, WEPD29.

[8] C. Thomsen, H. T. Grahn, H. J. Maris, and J. Tauc, "Surface generation and detection of phonons by picosecond light pulses," *Phys. Rev. B*, vol. 34, pp. 4129–4138, 1986.

[9] I. Bahns *et al.*, "Thermal and mechanical stability of Bragg reflectors for an XFELP." presented at FEL'17, Santa Fe, USA, TUC02, this conference.

[10] Y. V. Shvyd'ko, S. Stoupin, A. Cunsolo, A. H. Said, and X. Huang, "High-reflectivity high-resolution x-ray crystal optics with diamonds," *Nature Physics*, vol. 6, pp. 196–199, Jan 2010.

[11] S. Stoupin and Y. V. Shvyd'ko, "Thermal expansion of diamond at low temperatures," *Phys. Rev. Lett.*, vol. 104, p. 085901, Feb 2010.

[12] J.-H. Rhew and M. S. Lundstrom, "Drift-diffusion equation for ballistic transport in nanoscale metal-oxide-semiconductor field effect transistors," *Journal of Applied Physics*, vol. 92, no. 9, pp. 5196–5202, 2002.

[13] J. Maassen and M. Lundstrom, "A simple Boltzmann transport equation for ballistic to diffusive transient heat transport," *Journal of Applied Physics*, vol. 117, no. 13, p. 135102, 2015.

[14] J. Maassen and M. Lundstrom, "Steady-state heat transport: Ballistic-to-diffusive with Fourier's law," *Journal of Applied Physics*, vol. 117, no. 3, p. 035104, 2015.

[15] D. P. Sellan, J. E. Turney, A. J. H. McGaughey, and C. H. Amon, "Cross-plane phonon transport in thin films," *Journal of Applied Physics*, vol. 108, no. 11, p. 113524, 2010.

[16] G. Chen, "Thermal conductivity and ballistic-phonon transport in the cross-plane direction of superlattices," *Physical Review B*, vol. 57, no. 23, p. 14958, 1998.

[17] M. Farzaneh *et al.*, "Ccd-based thermoreflectance microscopy: principles and applications," *Journal of Physics D: Applied Physics*, vol. 42, no. 14, p. 143001, 2009.

[18] *The element six CVD diamond handbook*, Element Six Technologies US Corporation.