

Ultrafast x-ray thermometry: Contrasting strain and Debye-Waller effects in platinum thin filmsJ.-E. Pudell^{1,*}, M. Mattern^{2,†}, F. Baltrusch³, F. L. Boariu⁴, M. Kronseder⁵, R. Shayduk¹, A. Madsen¹, M. Rössle⁴, M. Bargheer^{3,4}, D. Schick², and A. von Reppert³¹*European X-ray Free-Electron Laser Facility, 22869 Schenefeld, Germany*²*Max Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, 12489 Berlin, Germany*³*Institut für Physik und Astronomie, Universität Potsdam, 14476 Potsdam, Germany*⁴*Helmholtz Zentrum Berlin, 12489 Berlin, Germany*⁵*Institut für Experimentelle und Angewandte Physik, Universität Regensburg, 93053 Regensburg, Germany*

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Thermal energy and temperature govern a wide range of physical properties and dynamics in solids. X-ray diffraction can be used to monitor material-specific temperatures on ultrafast timescales and within nanostructures by exploiting thermal expansion or the Debye-Waller effect. Here, we simultaneously track the intensity and position changes of different out-of-plane Bragg peaks of a 16 nm thick Pt film on an MgO substrate upon equilibrium heating and upon femtosecond laser excitation to quantify the mean-square atomic displacement and the lattice expansion. Our comparison of these two x-ray thermometers experimentally verifies that the in-plane expansion of homogeneously excited continuous single-crystalline thin films is forbidden on picosecond timescales. This drastically changes the out-of-plane thermal expansion coefficient, i.e., the relationship between the observed lattice expansion and the corresponding temperature increase. Thus, considering the boundary conditions of in-plane lattice expansion is generally indispensable for extracting temperatures from the lattice expansion in diffraction experiments.

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Introduction. The response of solids to ultrafast laser excitation is governed by the deposited energy and its spatiotemporal distribution [1–7]. Accordingly, energy and temperature are fundamental quantities in physical theories [8–13]. Developing these theories and identifying the role of laser-induced transient nonequilibria require quantitative access to the energy deposited in nanoscale layers on picosecond timescales.

Despite its crucial role, the deposited energy cannot be measured directly, as only its effects are experimentally accessible by probing the occupation of quantum states. Tracking the transient excitation of quasiparticles by spectroscopic [14,15] or scattering experiments [16–20] would be ideal for understanding the microscopic processes governing the ultrafast response of a material. However, this is experimentally demanding. In many situations, especially when the degrees of freedom are equilibrated, probing ensemble averages, such as determining a temperature change, suffices. Phonons, as the quantum excitations of the lattice, typically represent the largest energy reservoir, rendering them the ideal reference for the deposited energy. Diffraction experiments are a natural choice for probing phonon excitations through their

effect on the atomic lattice [16–18,21–31]. They provide a material-specific probe of heterostructures [25–27] and can be further focused to the nanoscale for imaging applications [5,7]. On the one hand, incoherently excited phonons reduce the intensity of Bragg peaks via the Debye-Waller effect [2,16,18,22,32]. On the other hand, their excitation within an anharmonic interatomic potential additionally changes the interatomic distance, as reflected by a shift of Bragg peaks in reciprocal space [33–36]. In contrast to the Debye-Waller effect, the lattice expansion might not only originate from phonon stress but also from electronic [28,37–39], magnetic [40–44], and even nonthermal stress contributions [33,45]. Thus, translating the transient strain to a transient temperature via the equilibrium expansion coefficient $\alpha(T)$ is not easily achievable in the case of long-lasting transient states, especially in the presence of phase transitions, which may introduce a nonlinear dependence on the deposited energy [41–43,46]. Furthermore, the comparison of the transient strain in laterally continuous and nanostructured films demonstrated a dependence of the observed strain on the sample morphology, further complicating the extraction of the transient temperature [41,47,48]. Here, we combine tracking the intensity and the shift of symmetric out-of-plane Bragg peaks as proxies for the deposited thermal energy into a 16 nm thick Pt layer grown on an MgO substrate at different pump-probe delays. The comparison reveals a marked discrepancy in the extracted temperature between the methods that use the Debye-Waller factor $B(T)$ and the linear expansion coefficient $\alpha(T)$ obtained under thermal equilibrium. This can be understood by noting that laterally uniform excitation of a continuous film generates symmetric in-plane stresses, which

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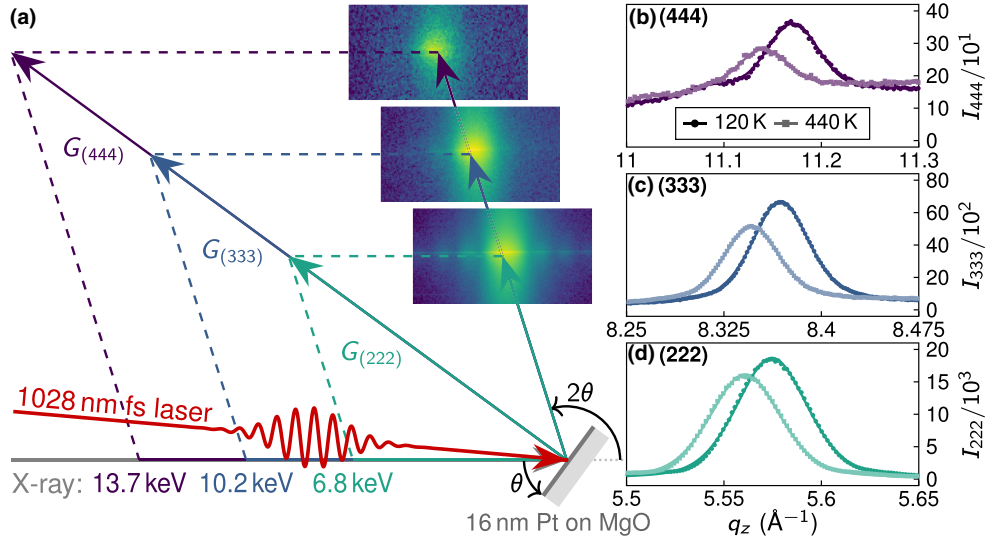


FIG. 1. Ultrafast x-ray diffraction experiment: (a) Sketch of the diffraction geometry probing the (222), (333), and (444) out-of-plane Bragg peaks of a 16 nm Pt (111)-oriented layer on MgO. Insets: reciprocal space maps recorded via symmetric $\theta - 2\theta$ scans. (b)–(d) $q_{x,y}$ -integrated Bragg peaks at 120 K and 440 K. The solid lines denote the fits of the Bragg peaks. Heating shifts the Bragg peaks to smaller reciprocal coordinates q_z (lattice expansion) and reduces their intensity (Debye-Waller effect).

restrict the material to expand only out of plane on ultrafast timescales. We find that the symmetry-forbidden in-plane expansion increases the ultrafast out-of-plane thermal expansion coefficient by a factor of 2.2 for (111)-oriented Pt via the Poisson effect, in perfect agreement with elastic theory. This highlights the need to generally consider in-plane lattice expansion when extracting temperatures from the out-of-plane lattice expansion.

Experiment. The ultrafast x-ray diffraction (UXRD) experiment was performed at XPP@KMC-3 endstation at the synchrotron source BESSY II [49]. A single-crystalline 16 nm thick (111)-oriented Pt film, grown by molecular beam epitaxy on an MgO(001) substrate, was either heated in equilibrium or excited by a 600 fs, 1028 nm pump pulse synchronized with the storage ring operated in single bunch mode. Details on the experiment and the structural characterization of the sample are provided in the Supplemental Material [50] (see also Refs. [49,51–69] therein). The static x-ray characterization identifies an in-plane coherence length of ≈ 200 nm and the density of the Pt film to be the same as bulk Pt. This highlights the high quality and the continuous, homogeneous character of the investigated Pt film confirmed by scanning electron microscopy measurements (see Fig. S7 [50]). Diffracted 80 ps x-ray pulses were recorded with a gated Pilatus 100 k detector [49]. To maintain identical pump conditions across diffraction orders, we accessed (222), (333), and (444) by changing the photon energy at a fixed scattering geometry [see Fig. 1(a)]. The position and intensity of the structural Pt Bragg peaks were tracked by reciprocal space maps (RSMs) recorded using $\theta - 2\theta$ scans around the Bragg angle of 53° with respect to the sample surface for photon energies of 6.8 keV, 10.2 keV, and 13.6 keV, respectively. Integration along the $q_{x,y}$ direction yields the diffractograms of the different diffraction orders shown for 120 K (dots) and 440 K (squares) in Figs. 1(b)–1(d). To complete the set of probed diffraction orders, we additionally track the peak posi-

tion and intensity of the (111) Bragg peak with 6 keV photons at the Bragg angle of 27.1° . We observe a decrease in the absolute intensity by approximately one order of magnitude with each diffraction order, which results in considerably different uncertainties in determining the position and intensity of the Bragg peaks. We extract the position Q_z and intensity I of the Bragg peaks by fitting the diffractogram along the out-of-plane reciprocal coordinate q_z with a pseudo-Voigt function and a linear background, which becomes increasingly relevant for higher diffraction orders.

Results. Using a static heating experiment, we characterize our sample and confirm the literature values for the temperature-dependent linear expansion coefficient $\alpha_{\text{Pt}}(T)$ [66] and Debye-Waller parameter $B_{\text{Pt}}(T)$ [68] in thermal equilibrium, thereby preparing for the time-resolved pump-probe measurements. We heat the sample from 120 K to 440 K and record RSMs around the (111), (222), (333), and (444) Bragg peaks every 20 K. The observed relative change in the out-of-plane Bragg peak position Q_z with respect to its position at 120 K defines the measured out-of-plane lattice strain $\eta_3(T) = -\frac{Q_z(T) - Q_z(120 \text{ K})}{Q_z(T)}$, where the index $i = 3$ denotes the out-of-plane direction. In general, the temperature-dependent out-of-plane strain $\eta_3(T)$ is given by

$$\eta_3(T) = \int_{T_0}^T \alpha_{3,\text{layer}}(T') dT' \quad (1)$$

with the reference temperature T_0 chosen to be 120 K and the out-of-plane linear thermal expansion coefficient $\alpha_{3,\text{layer}}$ of the layer. Figure 2(a) displays the measured $\eta_3(T)$ for all four orders (symbols) and compares it with models considering different expansion coefficients $\alpha_{3,\text{Pt}}(T)$. The gray dashed line corresponds to the literature expansion coefficient $\alpha_{3,\text{Pt}}(T)$ [66]. It does not reproduce the decreasing slope of the strain with increasing temperature. The experimental results are captured more accurately when assuming that the

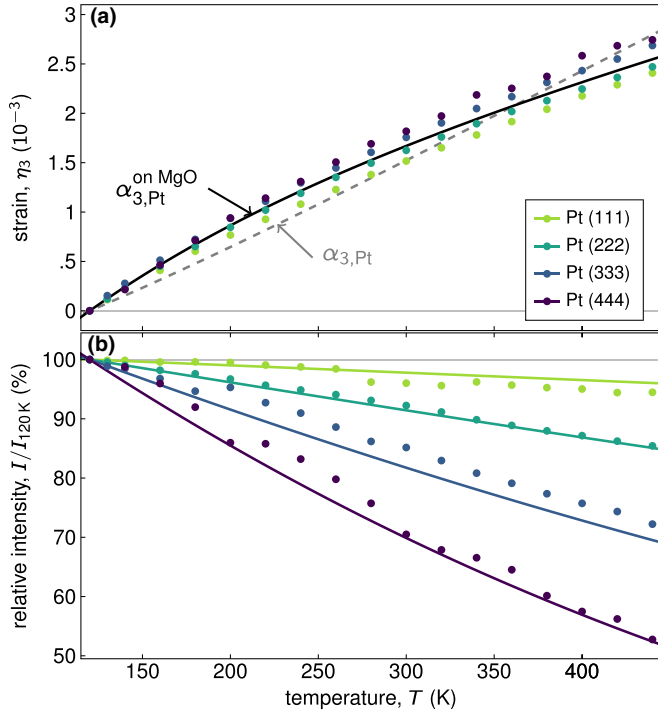


FIG. 2. Temperature-dependent strain and Bragg peak intensity: (a) Out-of-plane strain η_3 extracted from the Bragg peak shift with respect to the position at 120 K for all four diffraction orders (symbols). Lines denote the strain given by Eq. (1) with the linear thermal expansion coefficient $\alpha_{3,\text{Pt}}$ of bulk Pt (dashed, gray) and of Pt clamped to the MgO substrate $\alpha_{3,\text{Pt}}^{\text{on MgO}}$ (solid, black) defined by Eq. (3). (b) Relative intensity $I/I_{120\text{K}}$ of all four diffraction orders. Lines denote the calculation of the Debye-Waller effect [Eq. (5)].

temperature-dependent in-plane expansion of the Pt layer is dictated by the MgO substrate due to epitaxial clamping (black line). In this case, the out-of-plane expansion coefficient $\alpha_{3,\text{layer}}^{\text{on sub}}(T)$ of the layer on a substrate is generally given by [36]

$$\alpha_{3,\text{layer}}^{\text{on sub}}(T) = \alpha_{3,\text{layer}}(T) + \sum_{i \neq 3} \frac{c_{i3}}{c_{33}} (\alpha_{i,\text{layer}}(T) - \alpha_{i,\text{sub}}(T)) \quad (2)$$

with the direction dependent expansion coefficient of the substrate $\alpha_{i,\text{sub}}(T)$ and the elastic tensor elements of the layer c_{i3} in Voigt notation. In the case of Pt and MgO, the isotropic expansion and cubic lattice symmetry simplify Eq. (2) by $\alpha_i = \alpha_3$ and $c_{13} = c_{23}$. The out-of-plane expansion coefficient of Pt on MgO is

$$\alpha_{3,\text{Pt}}^{\text{on MgO}}(T) = \alpha_{3,\text{Pt}}(T) + 2 \frac{c_{13}}{c_{33}} (\alpha_{3,\text{Pt}}(T) - \alpha_{3,\text{MgO}}(T)) \quad (3)$$

with the elastic tensor elements in the (111)-oriented coordinate system c_{13} and c_{33} , and the bulk thermal expansion coefficient $\alpha_{3,\text{MgO}}(T)$ of MgO. Alternatively, Eq. (3) can be rewritten for isotropic materials using the Poisson factor ν [31,70]:

$$\alpha_{3,\text{Pt}}^{\text{on MgO}}(T) = \alpha_{3,\text{Pt}}(T) + \frac{2\nu_{\text{Pt}}}{1 - \nu_{\text{Pt}}} (\alpha_{3,\text{Pt}}(T) - \alpha_{3,\text{MgO}}(T)). \quad (4)$$

Due to the different Debye temperatures of Pt (230 K) [71] and MgO (740 K) [72] and the resulting differences in the temperature dependencies of their thermal expansion coefficients, $\alpha_{3,\text{Pt}}^{\text{on MgO}}(T)$ differs from $\alpha_{3,\text{Pt}}(T)$. Nonetheless, the correction is rather small due to the similar thermal expansion coefficients of Pt and MgO above 300 K.

Figure 2(b) displays the corresponding relative intensity $I/I_{120\text{K}}$ of the Bragg peaks with respect to their intensity $I_{120\text{K}}$ at 120 K (symbols). The solid lines denote the calculated intensities for the different diffraction orders,

$$I(T) = I_{\text{ref}} \exp(-Q^2 \langle u^2(T) \rangle), \quad (5)$$

with the reference intensity I_{ref} , the scattering vector Q , and the temperature-dependent mean squared atomic displacement $\langle u^2(T) \rangle$ described by the Debye-Waller parameter $B(T)$ via $\langle u^2(T) \rangle = \frac{B(T)}{8\pi^2}$. Our calculation based on the Debye-Waller parameter $B(T)$ reported in the literature [68] matches the intensity of all four diffraction orders.

For the time-resolved experiment, we used a subpicosecond laser beam with diameters of 750 μm and 780 μm (FWHM), incident at 55° with respect to the sample surface, nearly parallel (2° offset) to the x-ray beam, which has approximately half the size (250 μm and 500 μm) to ensure laterally homogeneous excitation of the probed volume. The laser's pulse energy of 115 μJ at a repetition rate of 104 kHz causes quasistatic heating of the Pt layer to 450 K relative to the set temperature of 330 K of the sample holder due to continuous energy deposition. This average base temperature in the pump-probe experiment is inferred from the laser-induced peak shift of the (111) Bragg peak by comparison with Fig. 2(a) at negative time delay, where the x-rays probe the film immediately before the next laser excitation. Note, the expansion induced by this quasistatic laser heating is characterized by the same expansion coefficient $\alpha_{3,\text{Pt}}^{\text{on MgO}}$ as the equilibrium heating in Fig. 2.

Figure 3 displays the temporal evolution of the Bragg peak parameters upon photoexcitation of the Pt layer relative to the base temperature of 450 K. The (222), (333), and (444) Bragg peaks were recorded under identical excitation conditions with an incident laser fluence of 15 mJ cm^{-2} . Since we measured the (111) Bragg peak at a smaller Bragg angle, we increased the incident fluence of the pump beam until achieving the same laser-induced lattice expansion. Upon laser excitation, we observe a rapid out-of-plane expansion of the Pt layer (positive strain) and a rapid decrease in the intensity of all Bragg diffraction orders. In order to compensate for changes in the incoming x-ray flux caused by long-term drifts of the beamline optics, we alternately measured time delays before and after optical excitation. The relative intensity $I(t)/I_0$ for each time delay can therefore be related to the average base intensity I_0 shortly before and after each data point. Our temporal resolution of about 80 ps is much longer than both the electron-phonon coupling time in Pt, approximately 2 ps [27,60], and the ~ 4 ps required for lattice expansion to propagate across the 16 nm film at the speed of sound [36]. Consequently, our measurements are insensitive to regimes in which electron pressure induces lattice expansion before substantial energy transfer to the phonon system occurs, as well as to scenarios in which coherently

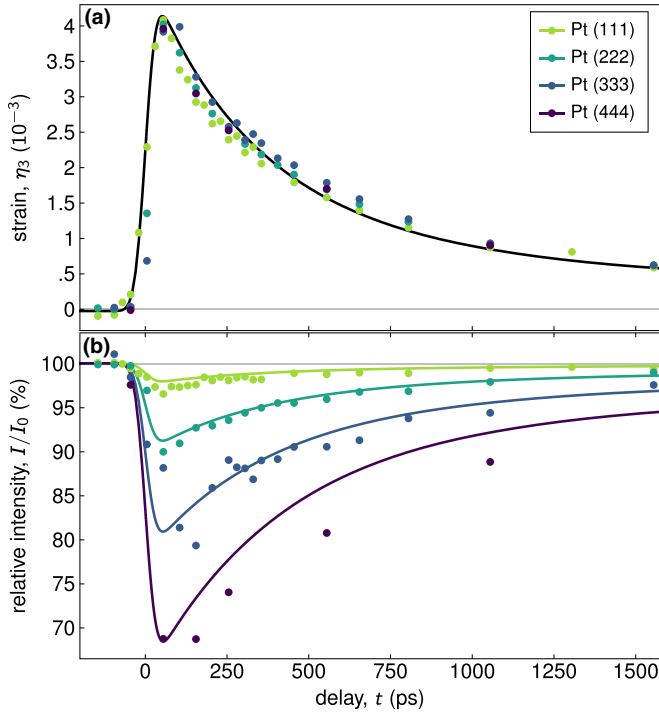


FIG. 3. Time-dependent strain and Bragg peak intensity: (a) Strain (symbols) after excitation at an incident fluence of 15 mJ cm^{-2} of all four diffraction orders. (b) Relative Bragg peak intensity (symbols) of all four diffraction orders. Lines denote the modeled laser-induced shift and relative intensity of Bragg peaks in the framework of dynamical x-ray scattering calculations.

driven strain pulses provide an additional contribution to the layer strain. By restricting ourselves to conditions where the phonon thermal energy predominantly governs both the strain and the Debye-Waller effect, we expect these two observables to yield identical temperatures when evaluated using Eq. (1) and Eq. (5). On longer timescales, the diffusive heat transport

via phonons from the photoexcited Pt layer into the MgO substrate results in a decrease in temperature. This is evidenced by the decreasing strain and the recovery of the Bragg peak intensity to the level prior to excitation on a timescale of hundreds of picoseconds.

Discussion. Figure 4 shows the strain η_3 derived from the Bragg peak shift as a function of the Bragg peak relative intensity I/I_0 in a scatter plot for all static temperatures from Fig. 2 (green circles) and for all time delays from Fig. 3 (blue squares). The colored solid lines denote the relationship between strain and intensity numerically calculated via Eq. (5) and Eq. (1) when considering different expansion coefficients: (i) of bulk Pt $\alpha_{3,\text{Pt}}(T)$ (light green), (ii) of Pt laterally clamped to an MgO substrate $\alpha_{3,\text{Pt}}^{\text{on MgO}}(T)$ (green), and (iii) the ultrafast expansion coefficient of Pt $\alpha_{3,\text{Pt}}^{\text{uf}}(T)$ (blue). The static experiment (green symbols) is well reproduced by $\alpha_{3,\text{Pt}}^{\text{on MgO}}(T)$ as given by Eq. (3). In fact, the total quadratic deviation χ^2 between the experiment and this model is statistically significantly smaller than the description by $\alpha_{3,\text{Pt}}(T)$ for the different orders. In contrast, the laser-induced dynamics (blue symbols) are not captured by $\alpha_{3,\text{Pt}}^{\text{on MgO}}(T)$ but only by the ultrafast expansion coefficient $\alpha_{3,\text{Pt}}^{\text{uf}}(T)$ (blue solid line). Its value can be derived by considering that the laterally homogeneous excitation of the probed volume results in a vanishing in-plane stress gradient, which suppresses any in-plane expansion on picosecond timescales [see inset in Fig. 4(d)]. Revisiting Eq. (2), this situation is comparable to the clamping of the film to a substrate without in-plane expansion

$$\alpha_{3,\text{layer}}^{\text{uf}}(T) = \sum_i \frac{c_{i3}}{c_{33}} \alpha_{i,\text{layer}}(T), \quad (6)$$

and specifically for Pt,

$$\begin{aligned} \alpha_{3,\text{Pt}}^{\text{uf}}(T) &= \left(1 + 2 \frac{c_{13}}{c_{33}}\right) \alpha_{3,\text{Pt}}(T) = \frac{1 + \nu_{\text{Pt}}}{1 - \nu_{\text{Pt}}} \alpha_{3,\text{Pt}}(T) \\ &= 2.2 \times \alpha_{3,\text{Pt}}(T) \end{aligned} \quad (7)$$

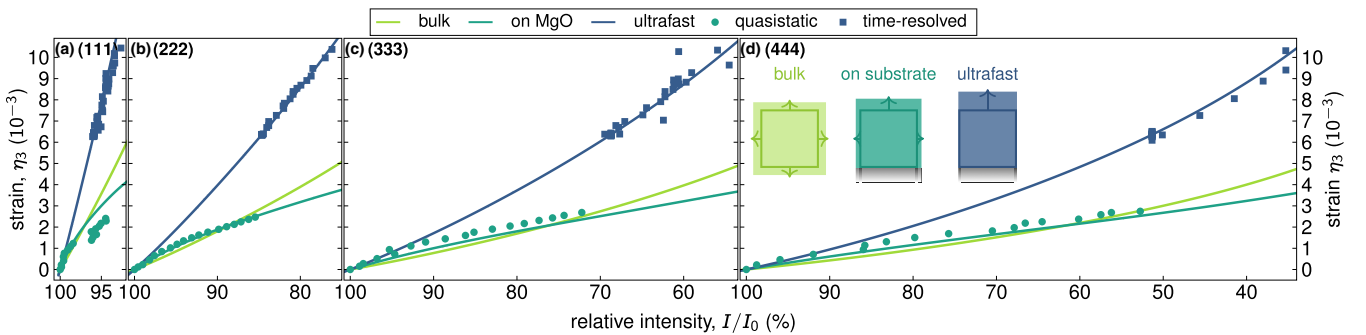


FIG. 4. Determination of the expansion coefficient from the strain-intensity relationship: Relative Bragg peak intensity I/I_0 vs strain η_3 with respect to the Bragg peak at 120 K of the data in Figs. 2 and 3 for the (a) (111), (b) (222), (c) (333), and (d) (444) Bragg peaks. UXRD results (blue squares) are best described by utilizing the ultrafast expansion coefficient $\alpha_{3,\text{Pt}}^{\text{uf}}(T)$ [Eq. (7)] (blue solid line). Results of equilibrium heating (green circles) are best described by $\alpha_{3,\text{Pt}}^{\text{on MgO}}(T)$ [Eq. (3)] (green solid line) and not by the bulk thermal expansion coefficient $\alpha_{3,\text{Pt}}(T)$ (light green solid line). Insets in (d) illustrate the different expansion of bulk material, thin film, and in an ultrafast case. The reference intensity of the time-resolved data prior to excitation was scaled by a constant factor, and the strain correspondingly offset to match the 120 K reference point of the ultrafast model (blue solid line). These adjustments account for long-term beamline drifts that prevent a direct quantitative comparison between equilibrium heating and ultrafast measurements, as beamline realignments were required between datasets. Crucially, these global corrections do not affect the observed difference in slopes of the intensity-strain relation.

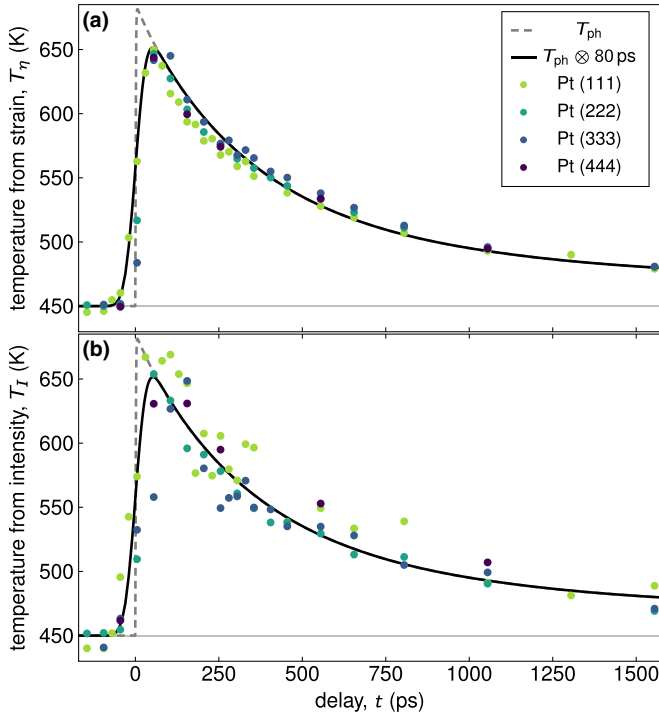


FIG. 5. Transient phonon temperature from the UXR experiment: Transient phonon temperature (symbols) extracted from (a) the transient strain and (b) relative Bragg peak intensities from the results in Fig. 3. Gray dashed line denotes the transient phonon temperature of the Pt layer modeled in the framework of a diffusive 2TM. Black solid line represents the convolution with an 80 ps instrument time resolution.

with c_{13} , c_{33} , and ν of Pt taken from the literature [54,55]. Note, in case of an isotropic material the ultrafast expansion coefficient [Eq. (7)] is well in line with the solution of the three-dimensional wave equation under the assumption of vanishing in-plane stress gradients [73].

Our results demonstrate that the expansion coefficients obtained under near-equilibrium heating conditions are not applicable to time-resolved experiments on ultrafast timescales. In particular, extracting a transient, layer-averaged temperature from picosecond strain measurements requires an explicit treatment of the vanishing in-plane expansion resulting from the laterally homogeneous laser excitation of the probed volume. However, taking into account the crosstalk of in- and out-of-plane expansion via the Poisson effect is relevant not only on ultrafast timescales but also in thermal equilibrium if nanoscale layers are epitaxially grown on substrates.

To directly extract ultrafast temperatures from the time-resolved strain and intensity, Eqs. (1) and (5) are used. The results are shown in Fig. 5 utilizing the ultrafast expansion coefficient $\alpha_{3,\text{Pt}}^{\text{uf}}(T)$ of Pt [Eq. (7)]. Using $\alpha_{3,\text{Pt}}^{\text{uf}}(T)$ we extract the same transient temperatures from the transient lattice expansion [Fig. 5(a)] and Bragg peak intensity [Fig. 5(b)] for all four diffraction orders. In our experiment, the temperature determination by strain fluctuates less than the data from a Debye-Waller analysis. The intensity of the (111) Bragg peak is the least reliable temperature proxy, as small intensity changes correspond to large uncertainties in the inferred temperature. This demonstrates that extracting transient tem-

peratures from Bragg peak intensities with small uncertainties requires access to higher diffraction orders with large reciprocal lattice vectors to ensure a strong modulation of the peak intensity with the rising temperatures.

Simulation. In the last step, we compare the experimentally determined temperature to the expected evolution of the average phonon temperature in the Pt layer in the framework of a diffusive two-temperature model (d2TM) that uses literature values for the thermophysical material parameters and an additional Pt-MgO interface conductivity of $200 \text{ MW K}^{-1} \text{ m}^{-2}$ [74]. The modular Python library UDKM1DSIM [69] is perfectly suited to support ultrafast x-ray thermometry experiments. It supports calculations of spatiotemporal temperatures in the framework of 2TM or even N-temperature models. Starting with the optical absorption calculated by a transfer matrix algorithm, we use a diffusive two-temperature model to calculate the spatio-temporal electron and phonon temperatures in the 16 nm Pt layer. Utilizing the corresponding thermoelastic stress, the spatiotemporal strain upon laser excitation is calculated by solving the one-dimensional linear elastic wave equation within the framework of a one-dimensional linear chain model of masses and springs. The employed thermophysical literature values including the ultrafast expansion coefficient $\alpha_{3,\text{Pt}}^{\text{uf}}$ are summarized in Table S1 in the Supplemental Material [50]. Finally, dynamical x-ray scattering calculations yield the transient layer-specific Bragg peaks. With a recent upgrade, the toolbox is now able to model the intensity changes using the calculated transient temperature and the Debye-Waller parameter $B(T)$ [75]. Thus, the modeled Bragg peak shift encoding the strain and the Bragg peak intensity encoding the phonon temperature can be directly compared to the experiment. Our modeled transient average phonon temperature (gray dashed line in Fig. 5) nicely matches the experimentally extracted temperatures if we include the finite time resolution of the experiment by convoluting the simulation results with an 80 ps full-width half-maximum Gaussian (black solid line). Accordingly, the modeled evolution of the Bragg peaks (solid lines) based on this transient average phonon temperature captures the experimental results (symbols) for all four diffraction orders (see Fig. 3).

Conclusion and Outlook. In summary, we determined the transient average temperature of a 16 nm thick Pt layer on an MgO substrate via the Debye-Waller effect and the out-of-plane strain upon laser excitation. We demonstrate the crucial role of considering the in-plane expansion for interpreting the experimental out-of-plane expansion in thermal equilibrium and on ultrafast timescales. Our results show clear experimental evidence for the vanishing in-plane expansion upon laser excitation on picosecond timescales due to laterally homogeneous excitation. Only on timescales of tens of nanoseconds for typical pump-spot diameters of hundreds of micrometers, the in-plane expansion launched at the edge of the pump spot will propagate at the speed of sound into the probed volume which relaxes the modification of the expansion coefficient on ultrafast timescales. This result is central to the usual approach of x-ray and electron diffraction thermometry, which extracts transient layer temperatures from the picosecond strain response.

In addition to this physical insight, our UXR experiment demonstrates that strain is the more reliable temperature

proxy compared to the intensity at a monochromatic x-ray source. Unlike the Bragg peak intensity, quantifying its shift is not affected by drifting incident x-ray flux, limited normalization schemes, and does not require access to higher, typically less-bright diffraction orders. Furthermore, quantifying the strain does not necessarily require angle or photon energy scans, making it more time efficient when employing reciprocal space slicing (RSS) [76] with position-sensitive high-resolution area detectors.

Pushing the combination of strain and the Debye-Waller effect to (sub)picosecond timescales accessible via free electron lasers (XFELs) could provide unique insights into laser-induced processes that introduce stresses on the lattice. While the Bragg peak intensity exclusively probes the excitation of phonons, the laser-induced strain can be driven by electronic or magnetostrictive stresses. If the coupling to the phonons is relatively slow, nonthermal effects can even dominate. In this case, a comparison of the transient phonon temperature extracted from the Debye-Waller effect and the total stress driving the strain response enables the identification of nonequilibria among the different degrees of freedom, providing unique microscopic insights into ultrafast physics.

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J.E.P. and A.v.R. supervised and coordinated the project. J.E.P. conceived the experiment. The XPP@KMC-3 instrument was operated by J.E.P., M.R., A.v.R., F.B., and M.M. M.K. produced and characterized the sample. J.E.P., M.R., A.v.R., F.B., and F.L.B. performed the data evaluation. J.E.P. conducted the simulations and modeling, for which A.v.R., F.B., and M.M. provided background. D.S. extended the UDKM1DSIM Python toolbox to model the Debye-Waller effect. M.B. and A.M. supported the research collaboration and provided project funding and oversight. J.E.P. and M.M. wrote the manuscript. All authors discussed the results and contributed to the manuscript.

Data availability. The data, analysis, and simulation scripts that support the findings of this article are openly available at [77]. The data analysis and simulation codes developed for this work utilize the publicly available nx5d repository for data analysis [51,52] and the udkm1dsim repository for the simulations [69,75,78,79].

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