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ABSTRACT

We conceive and construct an *on-site in situ* high-pressure time-resolved ultrafast optical spectroscopy instrument that facilitates ultrafast pump–probe dynamics measurements under high pressure conditions. We integrate an ultrafast pump–probe spectroscopy system with a diamond anvil cell (DAC) system. Significantly, both the DAC and the sample are fixed within the light path without motion and rotation throughout the whole ultrafast spectroscopy experiment, including tuning and calibrating the pressure. This instrument thus avoids introducing artifacts due to sample motion or rotation, enabling precision high-pressure ultrafast pump–probe dynamics investigations. As a demonstrating example, we compare the effect of *on-site in situ* conditions with *off-site in situ* conditions on the ultrafast dynamics of Sr_2IrO_4 under 0–44.5 GPa high pressure. Our data and analysis show that conventional possible artifacts are greatly reduced by using the *on-site in situ* layout. Our work helps the high-pressure ultrafast science investigation develop into a promising new area, which enables the exploration of nonequilibrium excited quantum states in the high-pressure regime.

I. INTRODUCTION

Time-resolved ultrafast spectroscopy has been widely employed in condensed matter physics (and other sciences) due to its unique advantages,[1–3] such as ultrahigh temporal resolution, capability of detecting excited-states above the Fermi levels, and generation of coherent boson excitations. It has been extensively used to investigate a wide-range of condensed matter physics, such as high-temperature superconductivity,[4,5] complex phase transition,[6,7] coupling between different degrees of freedom,[8,9] coherent control,[10–12] and laser-induced novel quantum states.[13–15] However, to date, ultrafast spectroscopy and its extensions are mainly used to investigate time-resolved physical properties of materials (or atoms, molecules, cells, etc.) at ambient pressure.

The high pressure technique has also been widely employed in condensed matter physics (and other scientific areas) investigations owing to its unique capability of tuning the electronic band structure by directly modifying the lattice constants. Such a relatively clean method of external control leads to novel phenomena in superconducting,[16] topological,[17] strongly-correlated,[18] thermoelectric,[19] and other materials. However, so far, most high pressure physics (and other sciences) mainly focuses on equilibrium states, and nonequilibrium state high pressure ultrafast dynamics is relatively rarely explored.

In recent years, efforts have been devoted to combining the two areas together, driven by the rich and deep fundamental scientific demands aforementioned. Challenges mainly come from the data reliability. Because ultrafast dynamics investigations are very delicate experiments and pressure change can also easily induce rich complex physics effects, maintaining reliable data is very crucial for the starting and development of such a cross-field high pressure ultrafast dynamics. Two aspects are crucial for achieving reliable data. One is the development of high pressure ultrafast pump–probe spectroscopy instruments. The other equally important aspect is

explicitly mentioning the condition for obtaining reliable data in such experiments, preferably setting it up as a standard description in the future works reporting the conditions to guarantee a precise and comparable data acquisition and thus to minimize possible misleading or misuse of relevant phrases.

Conventionally, it has already become a mature technology to combine static *single*-beam optical spectroscopy with high-pressure techniques to measure physical properties.[20,21] It is of convention that in such experiments, *in situ* denotes only that the sample is not taking out of the diamond anvil cell (DAC); it does not necessarily mean that the DAC remains unmoved within the light path. In fact, when tuning and calibrating the pressure, the DAC (with sample enclosed) is often taken outside of the light path. Hence, *in situ* experiments in this content do not guarantee that there is no sample motion or rotation relative to the light beam. In a latter paragraph, we will discuss why this could potentially introduce artifacts, especially for *two*-beam ultrafast pump–probe experiments.

In this work, we introduce *on-site in situ* experiments, whereby, based on *in situ* experiments, it is further required that both the DAC and the sample are not taken outside of the light path, even for tuning and calibrating the high pressure.[22] Thus, we propose a description protocol for carrying out two beam (pump–probe) high pressure ultrafast spectroscopy experiments. We construct an *on-site in situ* high-pressure time-resolved ultrafast pump–probe spectroscopy instrument, maintaining that both the DAC and the sample are not taken outside of the light path. Using this instrument, we detect the pressure-dependent ultrafast dynamics of Sr_2IrO_4 and compare the results between under *on-site in situ* and *off-site in situ* experimental conditions. The results show that the *on-site in situ* design excludes more potential artifacts caused by the sample spot-to-spot surface fluctuation and greatly enhances the reliability of the pressure-dependent ultrafast dynamics data.

II. *ON-SITE IN SITU* AS A STANDARD DESCRIPTION

A. *On-site in situ* condition

As aforementioned, in high pressure physics, the phrase we define, *on-site in situ* condition, is a narrower and more specific concept than the *in situ* condition. We justify why this *on-site* condition is important for high pressure ultrafast dynamics investigation. Compared with single-beam optical spectroscopy experiments (e.g., Raman spectroscopy, optical absorption spectroscopy, photoluminescence spectroscopy, and x-ray diffraction), two-beam optical spectroscopy experiments need precise spatial and temporal overlaps on the sample surface; hence, it is more sensitive to the motion of the laser spot on the sample surface, as well as the sample rotation. Hence, being *on-site* is very crucial for the time-resolved ultrafast pump–probe spectroscopy experiments. Otherwise, it is difficult to determine whether there are any artifacts introduced due to the motion and (or) rotation of the sample.

B. The essence of being *on-site in situ*

We illustrate the reasoning in Fig. 1. Real samples are usually bulk crystals or thin films. They are not perfect infinite crystals. It is common that there are cracks or layer boundaries or domain boundaries on the sample surfaces [Fig. 1(a)]. When the DAC and sample are taken outside of the light path and then put back, repositioning fluctuations are easy to occur, leading to relatively prominent in-plane shifts of the laser spot position. The laser spots on such imperfections will cause different scatterings and yield uneven quality and quantity of useful signals. Hence, the collected signals will very likely exhibit sharply different amplitudes and lifetimes. As a result, the transient differential reflectivity $\Delta R/R$ could be very different when the pump and probe beams are overlapped on a flat zone or an uneven zone. When taking this signal variation as a real signal driven by varying pressure, temperature, laser fluence, or other controlling factors, artifacts are introduced. In Fig. 1(b), when there are steps on the sample surfaces, the focusing spots of the laser pulses have different sizes than those on a different step. Slight off-focus, as well as worse spatial overlap, may occur for non-collinear geometries. Hence, the density of the photo-carriers and the corresponding signals could be different. In some situations, the smoothness and light scattering of different steps are also different. Note that repositioning fluctuations can also result in prominent sample position shift along the z-axis, leading to similar problems caused by the steps as well as rotations. In Fig. 1(c), for samples with uneven doping concentrations, scattered impurity states, or non-uniform defects, the photo-excited carriers have different densities and lifetimes when different regions of the sample are illuminated. In Fig. 1(d), for many

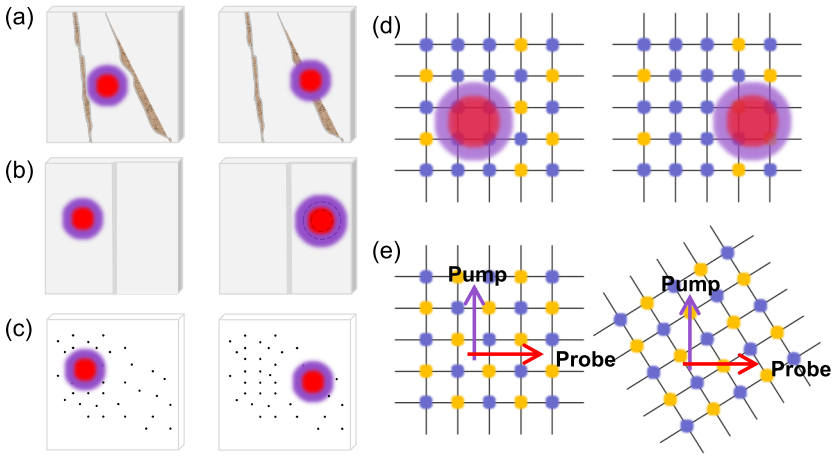

FIG. 1. Drawbacks of sample motion or rotation on obtaining reliable ultrafast pump–probe data: (a) when the sample surface has cracks or layer boundaries or domain boundaries; (b) when the sample surface has steps, the broken circles mark the laser spot sizes; (c) when there are non-uniform defect distributions or doping (vacancy) cites in the sample; (d) when the sample has in-plane superstructures or complex lattice structures; and (e) when the dynamics is sensitive to the orientation of the crystal lattice. Solid purple disk: 400 nm laser spot (pump beam). Solid red disk: 800 nm laser spot (probe beam).

samples with different in-plane superstructures or lattice structures, such as bulk FeSe, poly-crystals, and many other non-uniform crystals, the pump–probe signal is very sensitive to the laser spot locations. In Fig. 1(e), for samples sensitive to the polarizations of the excitation and detection pulses, the ultrafast spectroscopy signal is sensitive to the sample orientation (thus the sample rotation). In such a case, even for a perfect crystal (as frequently assumed in research works), rotation of samples leads to artifacts. Therefore, based on Figs. 1(a)–1(e), *on-site in situ* as a standard description is very necessary for a reliable data acquisition using high pressure ultrafast pump–probe spectroscopy instruments. Note that for ultrafast terahertz spectroscopy, because the laser spot is usually much larger than that of visible or near-infrared light, the requirement of the *on-site in situ* condition is alleviated.

C. Current situation of *on-site in situ* condition

So far, there have been a few pioneering innovations toward realizing time-resolved high pressure ultrafast pump–probe spectroscopy experiments. The majority employs DAC techniques[22–32] and a piston-cylinder design has also been reported.[33] More cases reported a pressure range at or below a few GPa, while less cases reported relatively higher pressure ranges up to a few tens of GPa. The maximum pressure value depends on the DAC technologies employed. Among these excellent attempts, however, only very few pioneering works[22,24,25] explicitly mention the *in situ* condition. In our recent work,[22] for the first time, we introduced the phrase *on-site in situ*. However, that work focuses on the experiments and results only, and we did not explain the important details about the instrument and the standard description there. Here, we further define the *off-site in situ* condition as that the sample remains in the DAC, but the DAC is taken outside of and put back into the light path during the experiment. Both *on-site* and *off-site* conditions could be classified as *in situ* conditions. It is necessary and essential to explicitly distinguish these two conditions. Hence, in this work, we propose making a precise classification by introducing the "*on-site in situ*" condition as a standard description for describing similar experiments. We note that in synchrotron beamline experiments, precise kinematic mounts are routinely employed, and thus, the experimental conditions are also guaranteed to be equivalent to "*on-site in situ*" conditions.

D. Challenges for realizing *on-site in situ* condition

The *on-site in situ* pressure-dependent ultrafast pump–probe dynamics investigation consists of three crucial functionalities: (1) measuring the relative differential reflectivity $\Delta R/R$ to obtain the ultrafast dynamics, (2) applying and tuning pressure on the sample, and (3) calibrating the pressure on the sample. These can be realized easily using the *off-site in situ* condition. However, it will potentially bring in large artifacts in a varying pressure experiment due to the sample motion or rotation.

Four major challenges are encountered to realize the *on-site in situ* condition: (1) Choose a type of high pressure technique such that when tuning and calibrating the high pressure, both the sample and the sample holder are not taken outside of the light path so that the sample's location and orientation are kept unchanged. (2) Because for higher pressure experiments the size of the diamond culet is small and hence the samples are small, microscopic spectroscopy is crucially required to achieve small enough laser spot size. This leads to both a weak signal from a small sample area and high risk of laser heating owing to a small sample volume. Note that this is an extra consideration compared with similar THz spectroscopies. (3) Furthermore, for such a microscopic spectroscopy setup, the pump and probe light pulses are usually aligned in a collinear way to achieve temporal and spatial overlap on the micron-sized sample surface. Hence, the pump and probe beams usually have to be of different central wavelengths to avoid interference. (4) Because the sample size is small, the sample vibrations and motions much be kept very small, leading to a stricter requirement for the *on-site in situ* condition.

III. INSTRUMENT DESIGN

Owing to the limited space in arranging the experimental elements in the setup, there are not many methods that can be used to tune and calibrate the high pressure in a way fulfilling the *on-site in situ* condition. We choose the DAC technique to realize the high pressure condition, and we use a membrane system for our DAC infrastructure to tune the pressure. We employ a collinear scheme, where the DAC is in a transmission geometry. Both the second harmonic generation method and an optical parametric amplifier are used to realize the different central wavelengths. We also discuss how to avoid laser heating and enhance the signal-to-noise ratio.

In Fig. 2, we schematically show the setup of our *on-site in situ* high-pressure ultrafast spectroscopy instrument. The time-resolved pump–probe light path is set to be horizontal, where reflection is detected from the front side. We apply and tune the high pressure by using a pneumatic membrane control system. The pressure is *on-site* calibrated and monitored from the back side of the DAC. This instrument consists of three major systems: (A) Time-resolved ultrafast pump–probe system. We construct a microscopic pump–probe system, whereby the time-resolved ultrafast pump–probe measurement is in a similar way as the conventional ones. (B) *On-site in situ* pressure tuning system. We apply pressure by using a commercially available pneumatic membrane control system (Druck PACE5000), whereby regular nitrogen gas is used to load pressure. (C) *On-site in situ* pressure calibration system. Back direction is implemented, whereby a 532 nm continuous wave laser beam incidents on ruby nearby the sample, and we measure its fluorescence spectrum to monitor the hydrostatic pressure on the sample. Note that the advantage is that all three composing systems can work independently in a compatible way, without affecting each other.

A. Time-resolved ultrafast pump–probe system

1. Microscopic ultrafast spectroscopy

The DAC technology limits the sample size to be from a few to hundreds of microns as usual. Thus, the microscopic ultrafast pump–probe spectroscopy system is needed. Ultrafast laser pulses from a Ti:sapphire amplifier with 800 nm central wavelength and 70 fs pulse duration rate are used to excite and detect the ultrafast dynamics of quasiparticles (QPs). The advantage of using an amplifier rather than an oscillator is that, with a repetition rate of 250 kHz, one can greatly reduce the laser heating while

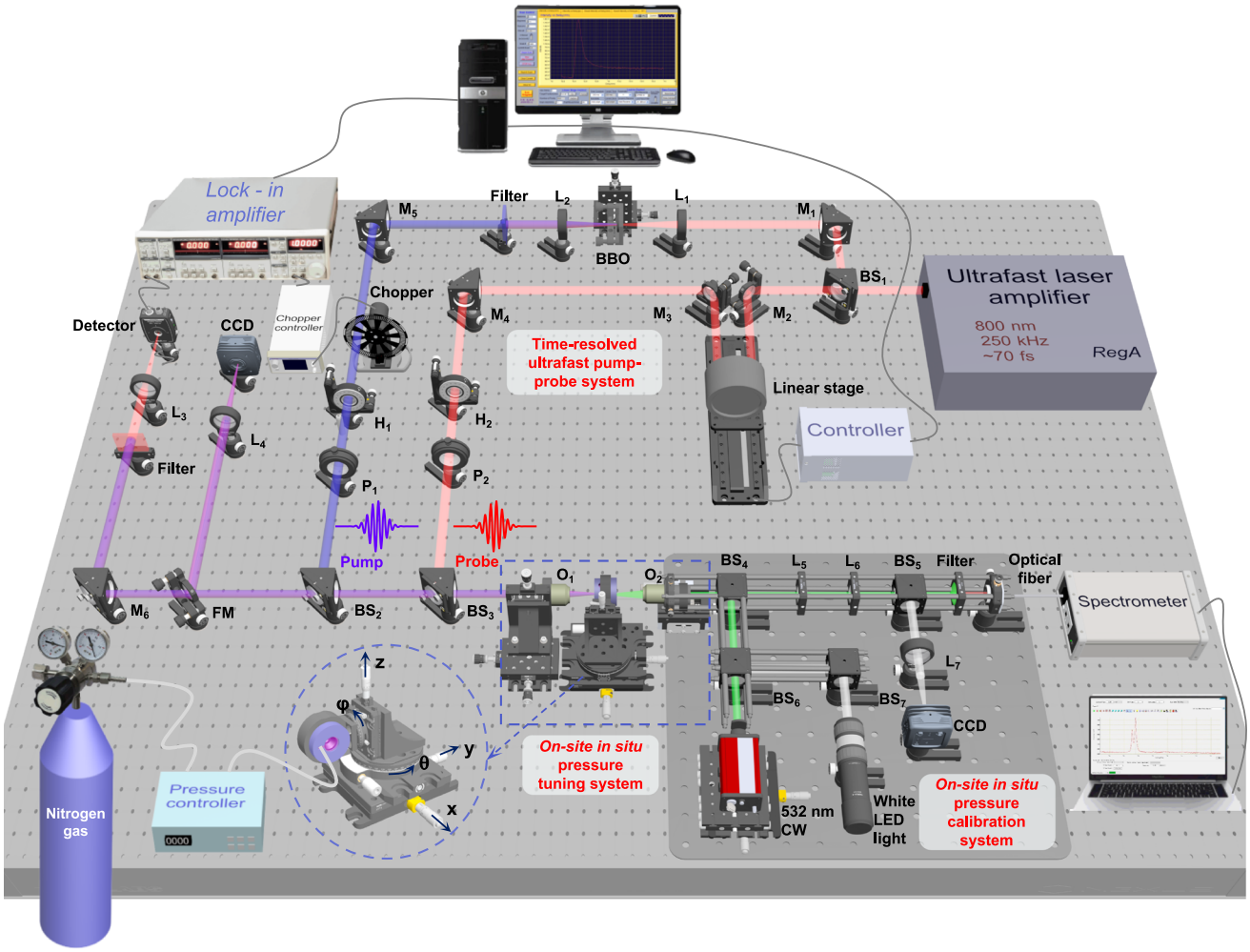

FIG. 2. Schematic experimental setup of the *on-site in situ* high-pressure time-resolved ultrafast spectroscopy instrument. This instrument consists of three systems: time-resolved ultrafast pump–probe system, *on-site in situ* pressure tuning system, and *on-site in situ* pressure calibration system. BS: beam splitter; H: half wavelength plate; P: polarizer; L: focusing lens; BBO: nonlinear crystal β-BaB_2O_4; FM: flip mirror; O: objective lens; and M: reflecting mirror. The sample stage is controllable for three translational dimensions and two rotational dimensions. The three systems can function independently in a compatible way.

maintaining good enough signal-to-noise ratio.[5,34,35] The time-resolved measurement is realized in a way similar to conventional pump–probe experiments.[4,5,36] Both beams are collinearly focused on the sample surface through an objective lens from the front direction of a DAC with normal incidence. The reflected beams traverse through the same objective lens, whereas the probe beam is collected by a photo-diode detector and the pump is blocked by a long-pass glass filter (Fig. 2). A CCD camera is used to monitor the beam overlap on the sample surface. To ensure a sturdy light path, we employ optical cage systems when needed to reduce the fluctuations of the laser spot location on the sample surface (not fully illustrated in Fig. 2). To facilitate the optical alignment, an adjustable stage with five degrees of freedom is used to hold the DAC (zoom-in inset of Fig. 2). The x, y, and z directions are adjusted for locating the sample position, and the θ and φ angles are adjusted for achieving normal reflections for the beams.

2. Tunable wavelength spectroscopy

We extend the wavelength of the pump beam to be tunable from 0.48 to 2.4 μm (0.52–2.59 eV) using an optical parametric amplifier (OPA9450, Coherent Inc.). Alternatively, a simpler way to generate different wavelengths is by using a β-BaB_2O_4 crystal to double the frequency. The former way is more advantageous because it can avoid damage to the diamonds. It is known that diamond exhibits very strong absorption at the ultraviolet wavelength and the absorption has a red shift[37] under high pressure (especially for those above 100 GPa).

B. *On-site in situ* pressure tuning system

The maximum achievable pressure is determined by multiple factors,[21] including the quality of the diamond, culet size, shape of the diamond, gasket materials, and sample quality. Figure 2

illustrates the schematics of the *on-site in situ* high-pressure tuning system and how it is integrated into the whole system. Figure 3 shows the details of the components. In Fig. 3(a), the ultrafast pump (400 nm) and probe (800 nm) pulses are illustrated, along with the continuous wave calibration (532 nm) beam. In Figs. 3(a) and 3(b), the components of the DAC are explicitly illustrated, with the relative positions of the sample, ruby, and gasket shown in the zoom-in view of the sample chamber. The DAC generates pressure by compressing the sample between two opposing diamond anvils with small culets and provides transparent windows for optical access. The culet size varies from tens of μm to a few mm depending on the target pressure and experimental conditions. The gasket is used to sustain a large pressure gradient and confine the sample and ruby in the chamber. The pressure transmitting medium (such as neon gas in our case) is used to create hydrostatic pressures. In Fig. 3(a), a 10× long working distance microscope lens [M Plan Apo near-infrared 10×] is used to enhance the working space for tuning pressure. Even after this improvement, there is only a distance less than a few millimeters between the objective lens and the DAC surface. One of the practical ways to realize pressure tuning in such a limited space is by using a pneumatic membrane system. In Fig. 3(c), the pneumatic membrane is shown, of which the white metal tube is connected to a nitrogen gas cylinder. By tuning the pressure controller, we are able to continuously and precisely tune the pressure in the pneumatic membrane, hence the pressure on the sample. Using this system, we have demonstrated ultrafast relaxation with an achievable high pressure tuning range of 0–44.5 GPa. We believe the limiting value can be easily further enhanced (for example, for fixed extremely high

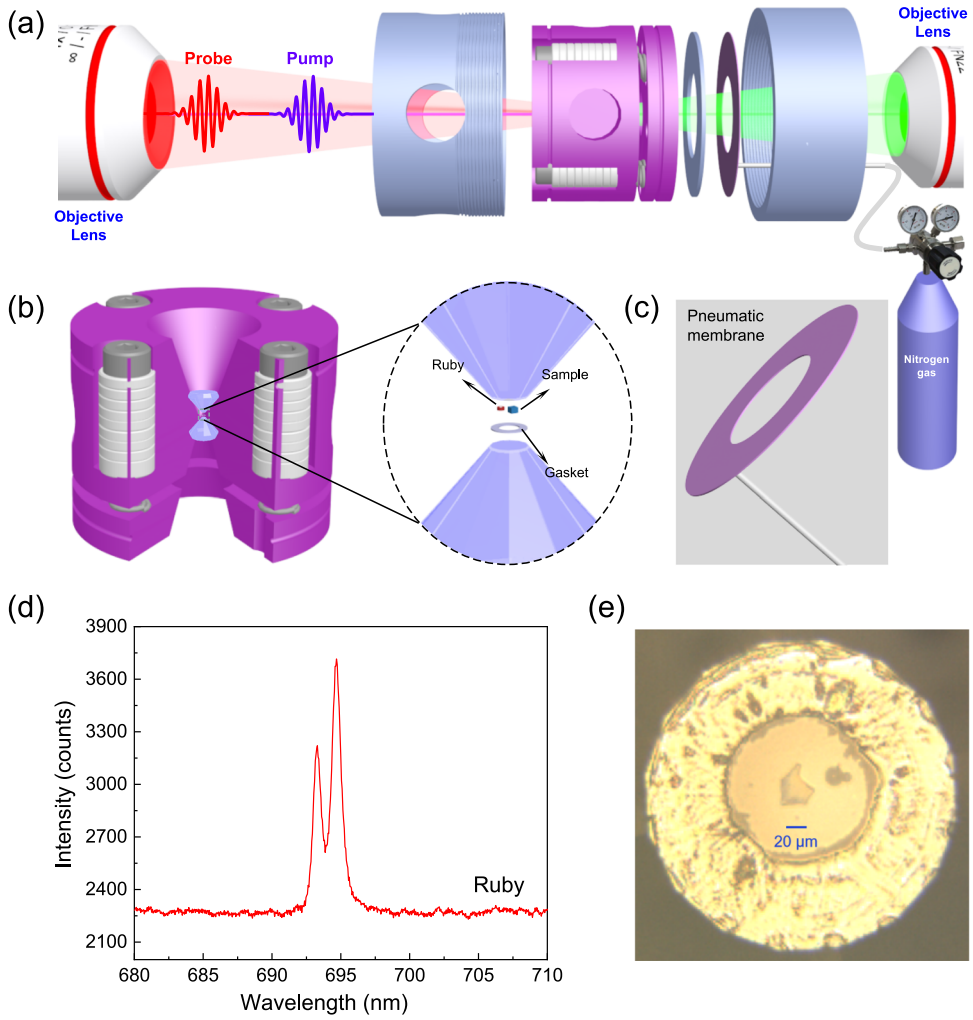

FIG. 3. (a) Schematic diagram of our design to realize the *on-site in situ* condition. The pneumatic membrane system is employed to tune the pressure on the sample. Pump and probe laser pulses are focused onto the sample surface from the front side of DAC, and a 532 nm cw laser beam is focused onto the ruby to calibrate the pressure from in a back direction geometry. (b) Schematic of the DAC. Zoom-in view: sample chamber. (c) Metallic pneumatic membrane for *on-site* controlling the pressure. White tube: metallic tube connected to the gas cylinder. (d) Characteristic fluorescence peaks of ruby. (e) Photograph of the sample and ruby taken by using our *on-site in situ* calibration system.

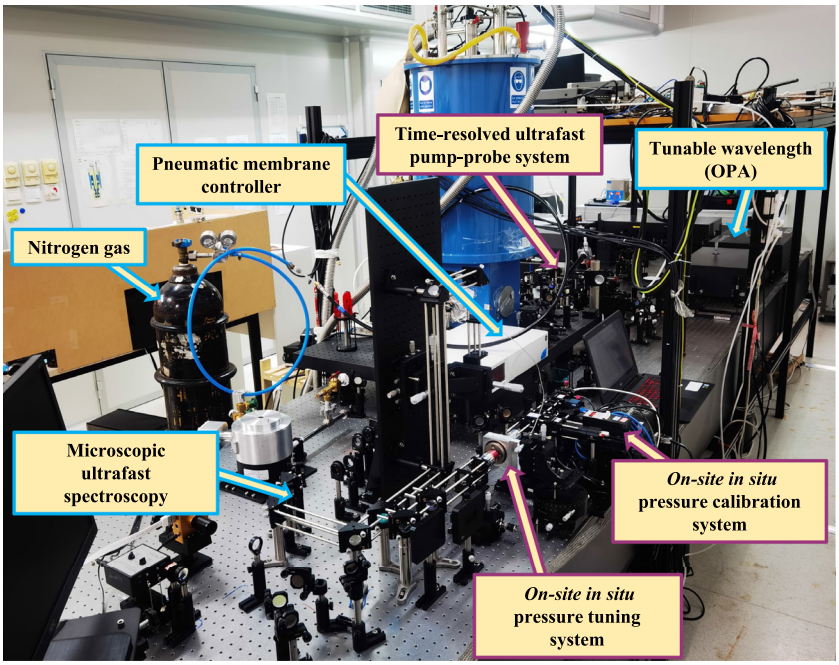

FIG. 4. Our *on-site in situ* high-pressure time-resolved ultrafast pump–probe spectroscopy instrument, which consists of three systems: time-resolved ultrafast pump-probe system, *on-site in situ* pressure tuning system, and *on-site in situ* pressure calibration system.

pressure experiments, we have tested obtaining ultrafast dynamics for a pressure value up to ~150 GPa).

C. *On-site in situ* pressure calibration system

We use ruby florescence shift (with its intensity and sharp florescence line) to calibrate the sample's pressure. In Figs. 2 and 3(a), the pressure calibration system is schematically shown. After going through a 10× objective lens, the reflected laser beam from the ruby is collected by using an optical fiber spectrometer, whereby the 532 nm excitation light beam is filtered out (Fig. 2). In Fig. 3(d), a typical characteristic florescence peak of ruby is illustrated, which corresponds to a specific pressure. Using the current ruby pressure calibration system, a maximum pressure of 80 GPa can be reached.[38] The flexible calibration system can also be used to monitor the morphology and position of the sample from the back direction of the DAC [e.g., with LED white light (Fig. 2)]. As an example, the morphology and location of the sample and ruby are clearly observed [Fig. 3(e)]. Like for the pressure tuning system, the *on-site in situ* condition is also implemented for the pressure calibration system. Because the three systems are independent to each other, the calibration is flexible, at any time needed. The same is true for monitoring the sample using this calibration system.

Overall, we show a photograph of our *on-site in situ* high-pressure time-resolved ultrafast spectroscopy instrument in Fig. 4 to give a real sense about the instrument. The crucial composing

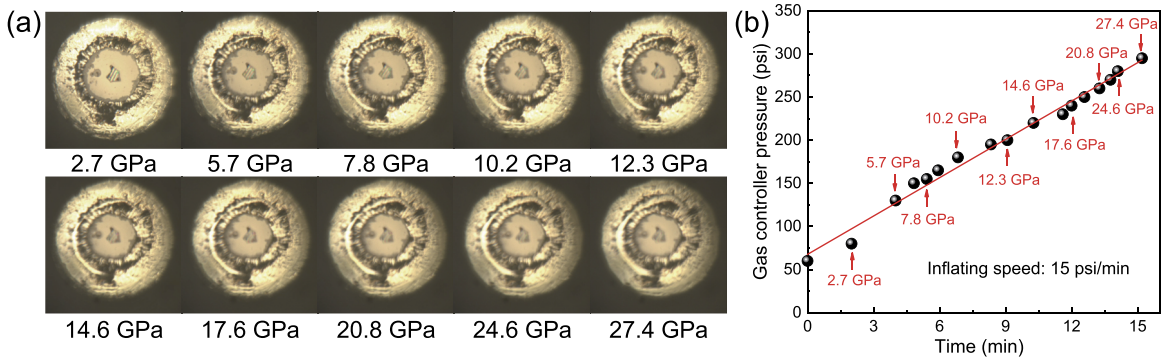

FIG. 5. (a) Photographs of a single crystal sample at several specific (increasing) pressures between 2.7 and 27.4 GPa tuned by a pneumatic membrane system. With increasing pressure, the DAC position has a slight motion of a few microns (see the text), which can be corrected by using the monitoring functionality in the *on-site in situ* pressure calibration system. (b) Controlling gas pressure in the membrane as a function of time (excluding the time for calibrating pressure). Red line: a fitting line to indicate the inflating speed, which is 15 psi/min. A high pressure tuning precision of 0.1 GPa is achieved under this inflating speed.

systems are marked directly on the photo. We have tested that this system can also be used for ultrafast pump–probe spectroscopy experiments at different temperature for samples in a DAC with a specific high pressure. We have also succeeded in implementing the system in a vertical light path geometry, which is in parallel to the horizontal light path geometry shown in Fig. 4. The OPA9450 system mentioned in Sec. III A 2 is also illustrated in Fig. 4.

IV. INSTRUMENT PERFORMANCE

We test the performance of this instrument fulfilling the *on-site in situ* condition. A sample of Sr_2IrO_4 (with a size of ~20 × 40 μm^2) is investigated, with an emphasis on comparing the results under *on-site in situ* and *off-site in situ* experimental conditions, respectively. The diamond culet diameter is 300 μm.

A. Monitoring the motion and rotation of the sample and DAC

For a pneumatic membrane system, at the initial stage when tuning up the pressure to a few GPa, it has been known that, as a common fact, the DAC position may slightly vary relative to the metallic embracing package due to the pressure gradient applied. This could be improved by developing more sophisticated high pressure DAC techniques. In Fig. 5(a), we show the photographs of the sample in the DAC with increasing pressure from 2.7 to 27.4 GPa. In this work, we observe an in-plane position change of the DAC up

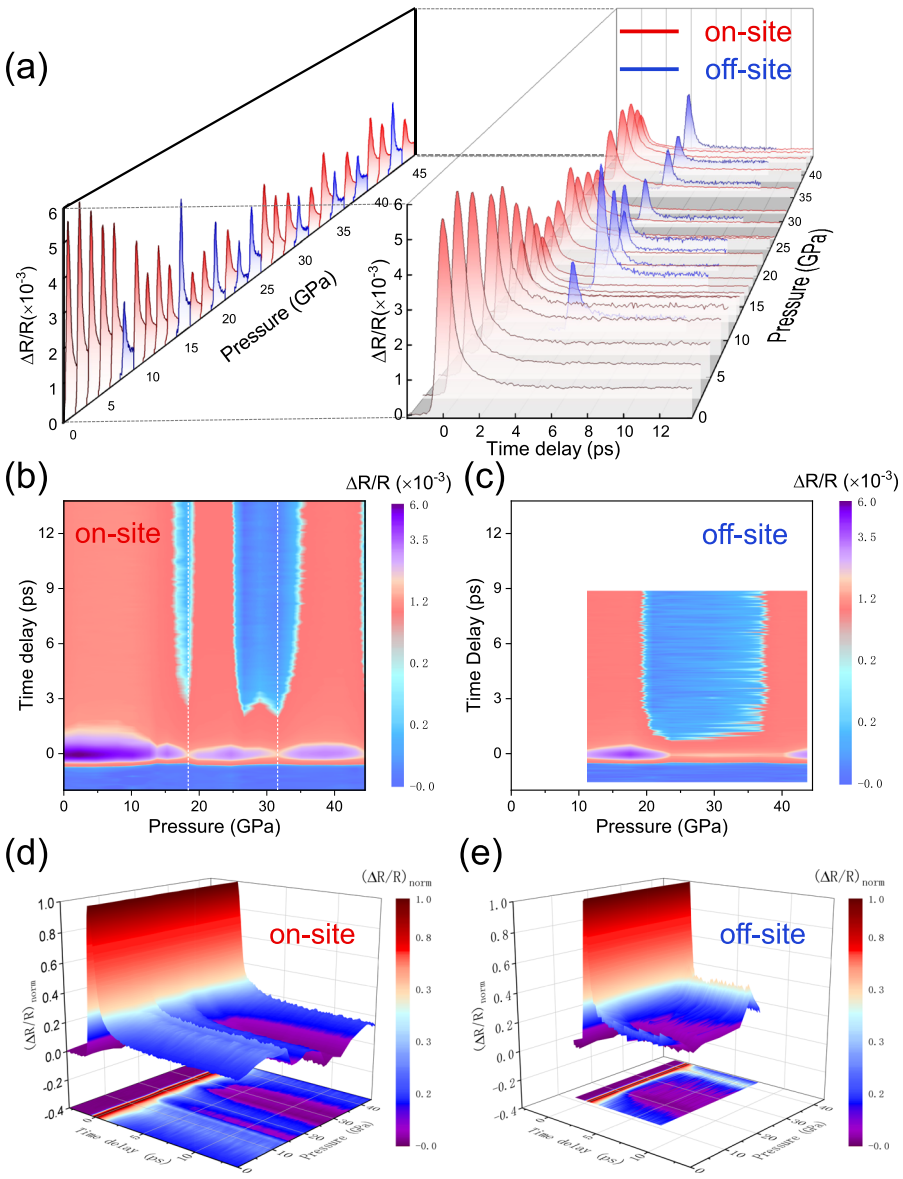

FIG. 6. (a) Comparison of the experimental data of differential reflectivity ΔR/R between *on-site in situ* and *off-site in situ* conditions. Red curves: data obtained with the *on-site in situ* condition (data adapted from Ref. 22). Blue curves: data obtained with the *off-site in situ* condition. Right panel: the blue curves are offset temporally for clarity. Left panel: projection of the data in the right panel. (b) and (c) 2D color map of the data in shown (a); adapted from Ref. 22. (d) and (e) 3D color map of the normalized data shown in (a).

to a few microns; at higher pressures above 5.7 GPa, no DAC position change is explicitly observed [Fig. 5(a)]. Thanks to our *on-site in situ* pressure calibration system, after tuning the pressure value, we are able to monitor the DAC location and manually tune the DAC back to its original position [not shown in Fig. 5(a)]. Motion along the z-direction is also observed; as shown in Fig. 5(a), the DAC image becomes a little unclear. The z-direction DAC motion can also be tuned back by monitoring the sample resolution using our *on-site in situ* pressure calibration system. In our experiment, at above 10.2 GPa, the z-direction DAC motion is so small that there is no need to adjust the DAC (back) at all. The most fluctuations that our instrument excludes are the repositioning fluctuations rather than the fine adjustments here. It is worthy to note that fixing the gas supplying metallic tube for the pneumatic membrane is crucial for removing another source of possible vibrations of the DAC, hence serious artifacts, in all related similar instruments and experiments.

We tune the gas pressure by the pneumatic membrane controller. The inflating gas pressure is set to increase 1 psi per step, ranging from 0 upto 295 psi. As a result, the overall inflating speed is 15 psi/min, which raises the pressure at a rate of 1.8 GPa/min [Fig. 5(b)]. Qualitatively, if a higher inflating speed is employed, the sample position will fluctuate with larger shifts. Here, we demonstrate an inflating speed at 15 psi/min or lower will be good enough [Fig. 5(a)]. However, further lowering the inflating speed will not definitely or prominently reduce the sample position fluctuation. This is related to the pneumatic membrane technology, of which we leave the detailed study to future investigations.

B. *On-site in situ* vs *off-site in situ* conditions: Experiments

We demonstrate the excellent performance of an *on-site in situ* system by comparing the results obtained using it with those using an *off-site in situ* system. The central wavelength of the probe beam is 800 nm and that of the pump is frequency-doubled to 400 nm by a β-BaB_2O_4 crystal, which is in line with the *on-site in situ* condition in Ref. 22. The fluences under the *off-site in situ* condition are kept as 5 and 1.8 mJ/cm^2, respectively. In Ref. 22, the pump and probe fluences under the *on-site in situ* condition are 2.66 and 0.45 mJ/cm^2, respectively. Investigations in Ref. 22 have shown that these fluence values are all within the linear range without thermal heating, and the differences between the values will not affect the comparison.

In the *off-site in situ* condition experiment, after achieving the ultrafast relaxation data at a specific pressure, we take the DAC out of the light path (i.e., *off-site*) to increase the pressure by mechanically tightening the screws of the DAC. Then, we calibrate the pressure value and put the DAC back into the light path. We follow the convention to keep the position and orientation of the sample as the same as those before taking out of the light path. We have a mark on the DAC stage and we also use a CCD camera to locate the sample. Even though with these efforts, experimental errors caused by resetting the sample location and orientation are likely generated for the *off-site in situ* experiments (see below sections).

C. *On-site in situ* vs *off-site in situ* conditions: Time-resolved data

In Fig. 6(a), we show the pressure-dependent ultrafast relaxation data of Sr_2IrO_4 at *on-site in situ* and *off-site in situ* conditions, respectively. The red curves represent *on-site in situ* data (data adapted from Ref. 22) and the blue curves are for *off-site in situ*. For clarity, the blue curves are all offset with an identical temporal interval. In the right panel of Fig. 6(a), the ultrafast QPs dynamics is prominently different for the *on-site in situ* and *off-site in situ* conditions. To see this more clearly, we project the data in the right panel onto the left panel. The amplitudes vary for the two sets of data. To see the difference between the two sets of data, we plot the 2D color mapping of the data in Figs. 6(b) and 6(c) (adapted from Ref. 22). Both the amplitudes and lifetimes are different, and a clear difference is identified for the light blue color region. The data under the *on-site* condition yield two separate pressure regions, while that of the *off-site* condition yield only one pressure region. We further illustrate the normalized $\Delta R/R$ data in a 3D color tomography form in Figs. 6(d) and 6(e). In such a way, the difference in lifetime is clearly seen, as reflected by the bottom projection patterns. Therefore, we conclude that the original ultrafast relaxation data themselves for the two different conditions are sharply different. As aforementioned in Sec. IV A, we have made sure that the *on-site in situ* data are reliable.

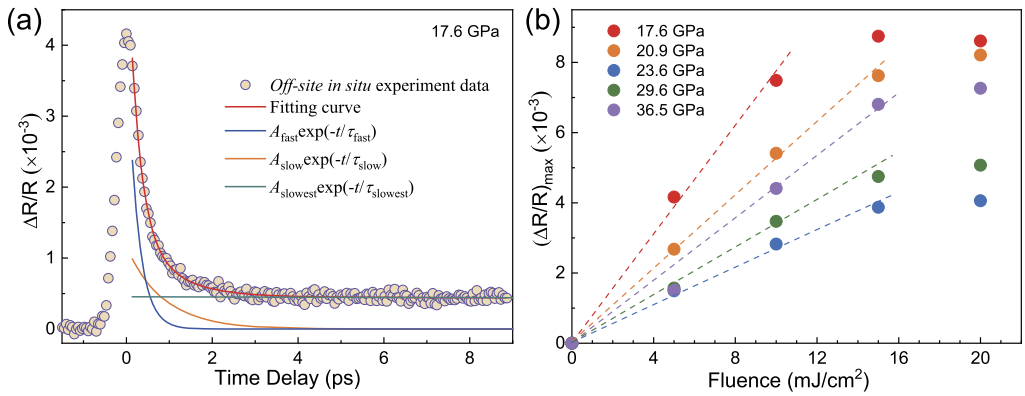

FIG. 7. (a) Data analysis of the $\Delta R/R$ signal for a typical scanning trace at the *off-site in situ* condition. Red curve: overall fitting curve, which is a sum of the three exponential decay functions represented by the blue, orange, and green curves. (b) Fluence dependence of $(\Delta R/R)_{max}$ at several pressure values. Dashed lines: linear relation is observed in the low-fluence regime.

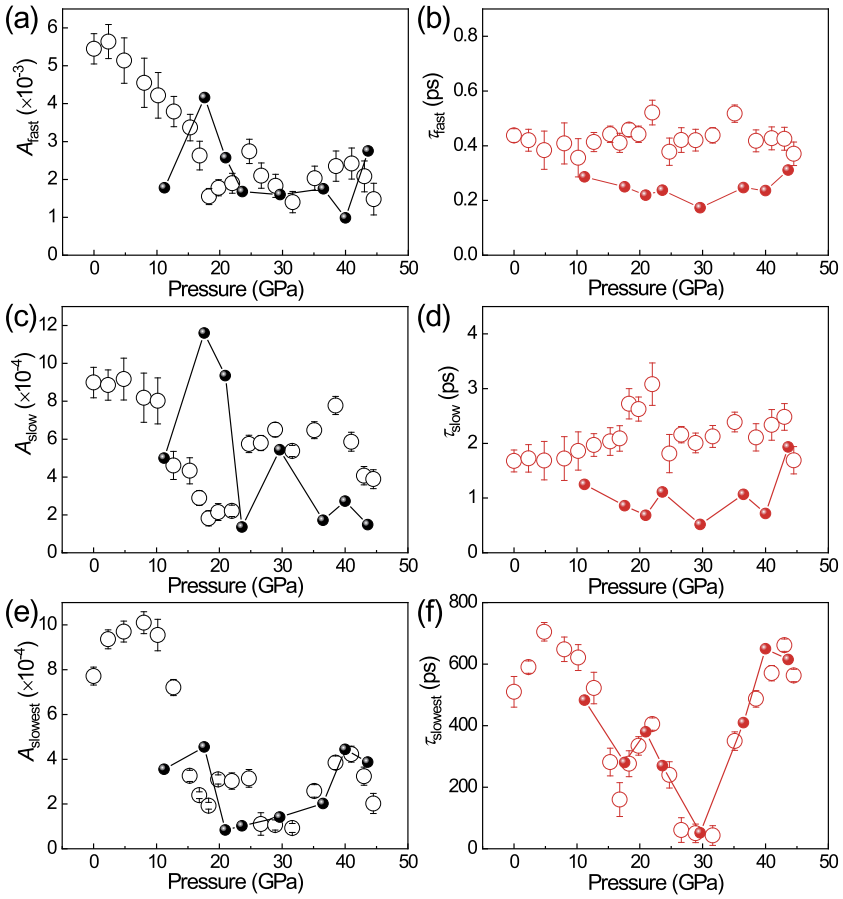

FIG. 8. Comparison of ultrafast dynamics (amplitude and lifetime) between *on-site in situ* and *off-site in situ* conditions. (a), (c), and (e) Pressure dependence of the amplitudes of the fast, slow, and slowest components. (b), (d), and (f) Pressure dependence of the lifetimes. Open circles: results with the *on-site in situ* condition (data adapted from Ref. 22). Solid spheres: results with the *off-site in situ* condition.

Thus, here we have demonstrated that the *off-site in situ* data indeed may contain artifacts due to taking the DAC and sample outside of the light path (which results in the motion or rotation of the sample).

D. *On-site in situ* vs *off-site in situ* conditions: Ultrafast dynamics

To quantitatively identify the difference for the two conditions, we further analyze the amplitudes and lifetimes of the ultrafast QPs relaxation data shown in Fig. 6. We fit each of the ΔR/R data using three exponential components. Because the data analysis for the *on-site in situ* condition has been given (see Fig. S2 of Ref. 22), here we only show the analysis for the *off-site in situ* condition. In Fig. 7(a), the overall ultrafast dynamics of Sr_2IrO_4 consists of three components (fast, slow, and slowest), each of which is explicitly shown. In Fig. 7(b), we also show the fluence dependence of the maximum |ΔR/R| value, which exhibits a linear relation for fluences below 10 mJ/cm^2. This indicates that at below 10 mJ/cm^2, the sample is not experiencing laser heating effects.[5,34,35]

We summarize the amplitudes and lifetimes of the three components in Fig. 8. The open circles denote ultrafast dynamics parameters corresponding to the *on-site in situ* condition (data adapted from Ref. 22), and the solid spheres denote those for *off-site in situ* condition. The values of the quantities in Figs. 8(a)–8(e) all manifest significant differences. Only the lifetime of the slowest component in Fig. 8(f) does not exhibit a prominent difference. The dynamics with the *on-site in situ* condition (slow component) exhibits a salient *pressure-induced phonon bottleneck effect* at 20 GPa,[22] whereas the dynamics with the *off-site in situ* condition (slow component) does not show such a feature. Therefore, we have demonstrated that intrinsic physical properties may be obscured by the potentially possible artifacts due to the motion and rotation of the samples under *off-site in situ* conditions. Hence, for a precision high pressure ultrafast dynamics investigation, it is preferred to employ an *on-site in situ* instrument to obtain reliable excited state physical properties.

V. SUMMARY

We propose and experimentally demonstrate the importance of implementing an *on-site in situ* condition for high-pressure ultrafast pump–probe dynamics experiments. We construct an *on-site in situ* high-pressure time-resolved ultrafast pump–probe spectroscopy (in the future, it may be abbreviated as HPUPS or HPUS or other similar forms) instrument. Both the sample and DAC are fixed in the light path during the whole ΔR/R experiment, especially during the tuning and calibration of the high pressure. We experimentally compare the results with *on-site in situ* vs *off-site in situ* conditions and evidence that using our constructed instrument with the *on-site*

in situ condition may potentially exclude the artifacts due to motions or rotations of the sample and DAC. Hence, the instrument we construct here contributes to obtaining reliable precision high pressure ultrafast dynamics data. This is crucial for excited state investigations in condensed matter physics under comprehensive extreme conditions. Our innovation thus also has wide outreaching impacts for other high pressure ultrafast science investigations.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Y.L.W. and X.Y. contributed equally to this work.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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