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Gold implanted germanium photoswitch for cavity dumping of a free-electron laser

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We present a plasma switch based on gold implanted germanium (Ge:Au) as a potential candidate for efficient cavity dumping of a free-electron laser (FEL). Ge:Au has a sub-nanosecond carrier lifetime – much shorter than the FEL pulsing period of 77 ns – and demonstrates a high photoinduced reflectivity in a broad range of infrared wavelengths from 6 to 90 μm . The Ge:Au plasma switch exhibits negligible absorption of the FEL radiation in the ‘off’ state and requires only moderate thermoelectric cooling for incident FEL power of several Watts. A reflectivity level of more than 50 % in the ‘on’ state is achieved over the entire spectral range of this study. The corresponding optical pump fluence exhibits a linear relationship with the FEL frequency. This scaling is corroborated by our simulations highlighting the role of a finite sub- μm thickness of the photoinduced reflecting plasma layer. The demonstrated device is promising for the realization of the FEL cavity dumping for experiments that simultaneously require higher pulse energy and lower average power.

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Free-electron lasers (FELs) are unique sources of intense narrowband terahertz (THz) radiation with a wide and continuous spectral tunability. The FELBE FEL located at the Helmholtz-Zentrum Dresden-Rossendorf^{1,2} is an oscillator type of FEL driven by a superconducting linear accelerator which operates in a continuous mode to achieve continuous lasing at a high pulse repetition rate of 13 MHz. This FEL has proven to be very useful for studying field-driven phenomena in bulk and nanostructured materials at photon energies between 5 and 250 meV (wavelengths from 250 to 5 μm , respectively).³⁻⁹ The high repetition rate and the high average power is ideal for a broad range of experimental techniques such as scanning scattering near-field microscopy,¹⁰⁻¹² time-resolved photoluminescence,¹³ single-color pump-probe measurements,¹⁴ and two-color pump-probe measurements using tabletop lasers synchronized to the FEL⁵. However, in experiments requiring the highest possible pulse energy, the resulting high average power due to the 13 MHz repetition rate can cause undesirable sample heating. This is especially critical for systems that must be studied at low temperatures, for example, superconductors or semiconductor nanostructures with low-energy electronic excitations or water-containing biological samples^{15,16}, that cannot withstand high heat loads.

Since the pulsing period of an oscillator type of FEL is defined by the cavity length and cannot be changed, the only possible way to increase this period is by selectively extracting pulses from the FEL cavity at a lower commensurate repetition rate using an appropriate optical switch. Especially advantageous is a scheme where the optical switch is located *inside* the FEL cavity and couples out the intracavity radiation as depicted in Figure 1(a). A semiconductor wafer is placed at Brewster's angle with respect to the cavity mode in order to minimize the cavity loss and to avoid outcoupling of the FEL pulses in the transparent state of the switch. A femtosecond laser synchronized to the FEL pulse train, but running at a much lower repetition rate excites an electron-hole plasma in the semiconductor wafer switching it from the transparent to a highly reflective state. The remaining weak intracavity pulse provides a seed pulse that is amplified during the following cavity round-trips until gain saturation when the nominal pulse energy is restored. This approach, which is often referred to as "cavity dumping", enables one to achieve an increased peak power, by selectively extracting a high percentage of the intracavity pulse energy, as opposed to the standard mode of operation where a few percent of the intracavity pulse energy is outcoupled on every pass.

The first practical realization of FEL cavity dumping was demonstrated for the millimeter-wave FEL at the University of California Santa Barbara (UCSB).¹⁷ Later on, this scheme was successfully implemented for pulsed electron paramagnetic resonance spectroscopy.¹⁸ The UCSB FEL operating at the emission frequency of 0.24 THz with a microsecond pulse duration demonstrated an 8-fold increase of peak power when being dumped using a Si-based photoswitch. However, such Si-based cavity dumper cannot be directly applied to the continuous pulsing mode of the FELBE FEL due to its slow recovery time. Several problems arise if the recovery time for the photoswitch is longer than the period between the FEL pulses. Firstly, subsequent residual FEL pulses will be also partially coupled out of the FEL cavity. Secondly, the incomplete recovery of the photoswitch transparency may suppress the FEL lasing. Furthermore, the residual absorption of the cavity dumper could lead to its damage by a thermal runaway. To achieve cavity dumping of a continuously pulsing FEL, a photoswitch must provide a high nominal transparency, and a fast switching from *off* (transparent) to *on* (reflective), and back to *off*. This is the necessary condition to ensure a stable operation of a FEL operating in the continuous pulsing mode. Considering the limiting factor of the round-trip time of the 13 MHz repetition rate of FELBE FEL cavity, we investigated materials for a switch with recovery time less than a few tens of ns.

Previously studied optical switches were based on polycrystalline Si,¹⁹ and Ge,²⁰ or more recently using single crystals of Si¹⁷ and Ge.²¹ Also, Ge has been used for sub-cycle slicing of high-field mid-infrared transients and can be useful for reducing the duration of FEL pulses.²² Both Si and Ge are non-polar semiconductors without phonon reststrahlen band and, therefore, they are ideally suited as cavity dumpers for FELs that operate in a very broad range of infrared frequencies. However, due to the indirect character of the bandgap in these materials the typical carrier lifetimes are very long (around few μ s depending on the carrier density).²³ Schmidt et al. demonstrated that the photoinduced reflectivity of a Ge switch can recover on a much faster timescale since the plasma frequency quickly drops below the FEL frequency.²¹ Therefore, it is possible to pick single pulses from the FEL pulse train with the repetition rate of 13 MHz. However, even though the reflectivity recovers on nanosecond timescale, the remaining free charge carriers in Ge/Si should make it opaque for the FEL radiation for much longer time.

The goal of the present work is to study reflectivity and transmission dynamics of Ge-based photoswitches *simultaneously* in order to examine their potential use as the FEL cavity

dumper. For these experiments we utilize radiation with different wavelengths ranging from 6 to 90 μm from the FELBE laser operating at its maximum power level. Furthermore, the FEL beam was focused to a spot size much smaller than the size of the transverse intracavity mode, thus, reaching pulse fluences similar to those inside the FEL cavity. Singh et al. have shown that a moderate gold (Au) doping of Ge can efficiently reduce the carrier lifetime²⁴, which we show leads to much faster transmission recovery times as compared to pure Ge. Thus, photoswitches based on Au implanted Ge wafers (Ge:Au) are expected to be perfect candidates for FEL cavity dumping.

The scheme of the experimental setup is shown in Fig. 1(b). The Ge or Ge:Au wafer with a thickness of 500 μm is mounted on a rotation stage, which is attached to an XYZ translation stage. The p-polarized FEL radiation outcoupled from the FEL cavity is focused onto the wafer using a spherical mirror with a focal length of 20 cm. The wafer is set at Brewster's angle of 76 degrees to the incoming FEL beam in order to suppress the reflection in the unexcited state of the photoswitch. The photoexcitation is driven by near-infrared pulses generated by a Ti:sapphire laser amplifier (Coherent Astrella) operating at a repetition rate of 1 kHz. The diameters of the FEL spot and the 1 kHz optical pump spot were approximately 1 mm and 2 mm, respectively. The near-infrared pulse train is synchronized to the FEL pulses at 13 MHz and can be delayed using a mechanical translation stage. The reflected and transmitted FEL beams are recollimated by parabolic mirror and focused on a FET-based THz detector with sub-ns response time.²⁵ The signal from the detector is examined and recorded using an oscilloscope with 1 GHz bandwidth (Keysight InfiniiVision DSOX4104A).

From the preliminary sample testing, we observed significant sample heating at full FEL power of 2.6 W that resulted in a strong transmission drop even without the photoexcitation. This gradual process that occurs on a timescale of several seconds is related to thermal runaway. Therefore, for proper functioning of the photoswitch the excess heat caused by the FEL radiation and the optical pump has to be taken out. We have determined that mounting the Ge wafer onto a copper plate attached to a thermoelectric Peltier element operating at 130 mW of the cooling power is sufficient for a stable operation of the photoswitch. A gold mirror mounted adjacent to the Ge wafer served as a reflectivity reference.

Fig. 2 shows the operation of a pure Ge wafer for 8 μm FEL wavelength and 10 mJ/cm^2 pump fluence. The top panel (a) shows the FEL pulse train with the period of 77 ns. In

agreement with a previous report,²¹ the reflected signal is dominated by a single FEL pulse picked by the Ge photoswitch immediately after the pump pulse as can be seen in Fig. 2(b). Although the following pulse is still partially reflected due to free charge carriers remaining 77 ns after photoexcitation, an even more drastic effect can be observed for the transmitted signal, which is shown in Fig. 2(c). As expected, the transmission is strongly suppressed immediately after the pump pulse. However, it does not recover completely after 77 ns. The amplitude of the following pulse is 5 times smaller than the amplitude before the dumping event. Thus, the FEL cavity loss from the cavity dumper would be around 80%, which would suppress the recovery to saturated lasing. This evidences that a photoconductive switch based on pure Ge is not suitable as a cavity dumper for an FEL operating in a continuous mode.

In order to achieve the desired performance of the Ge cavity dumper, the carrier lifetime has to be significantly reduced. Recently, it has been demonstrated that moderate gold doping reduces the lifetime to few ns – far below the FEL pulsing period of 77 ns.²⁴ The gold impurities form deep levels within the bandgap with a large capture cross-section, thereby reducing the carrier lifetime.^{26,27} In order to fabricate the Ge:Au sample, a nominally pure Ge wafer has been implanted by Au ions with an energy of 330 keV and a dose of 7×10^{12} ions/cm². Subsequently, the sample has been annealed in vacuum at 900 °C for 10 hours in order to diffuse implanted ions into the depth of the sample. This results in the gold density of about 4×10^{14} ions/cm³ near the surface of the implanted Ge wafer.²⁴ Even such a low concentration of the deep traps leads to a sufficient reduction of the carrier lifetime to a sub-ns level. At the same time, the dark resistivity of the Ge:Au remains high enough to ensure excellent transparency for the FEL radiation. The operation of the Ge:Au photoswitch at a repetition rate of 1 kHz is demonstrated in Fig. 3. A trace of the transmitted FEL pulse train at 13 MHz repetition rate and a wavelength of 6.3 μ m is shown in Fig. 3(a) without any optical pumping. The sample is switched to the reflective state by the pump pulse around the time of 80 ns. The reflected FEL pulses are shown in Fig. 3 (b). Although a single pulse dominates the signal, one can see that for later delays weak residual pulses are also present. Nevertheless, the relative ratio of the residual pulse versus the picked pulses is 0.3 % [see inset of Fig. 3(b)], much less than that for pure Ge switch shown in figure 2(b) and reported by Schmidt et al..²¹ This attests for a superior performance of the Ge:Au for the FEL pulse picking.

More importantly, the Ge:Au switch demonstrates a dramatic improvement in the transmittance recovery. The transmitted signal in the presence of the optical pump pulse at 80 ns is shown in Fig. 2(c). As expected, the amplitude of the transmitted pulse after the dumping event is strongly suppressed. However, the subsequent FEL pulses show complete recovery to the nominal transmission. It is noteworthy that the transmitted pulses before and after the picked pulse have nearly the same magnitude (see Fig. 3(c)), indicating that the photoswitch returns to its initial state within the FEL pulsing period of 77 ns.

In order to obtain a more detailed understanding of the temporal behavior, we also recorded the reflectivity dynamics of the Ge:Au switch by measuring near-infrared pump / FEL probe traces for different FEL wavelengths. Fig. 4(a) shows the relative photoinduced reflectivity changes ($\Delta R/R_{\text{Gold}}$) for selected pump fluences as a function of time delay between FEL pulses (with the wavelength of 8 μm) and the pump pulses. A rise time of 20 ps, may be seen in Fig 4 (a) and is determined by the tilt between the FEL and the optical beam. The recovery time depends on the pump fluence, but even for the highest pump fluence of 10 mJ/cm^2 the reflectivity drops to nearly zero within 300 ps, thus confirming the sub-ns carrier lifetime. A similar sub-ns recovery for the Ge:Au sample has been observed for other FEL wavelengths of 6, 8 and 10 μm .

Another important parameter is the optical pump fluence (Φ_c) sufficient for efficient FEL cavity dumping (at least 50%) at a given wavelength. Figure 4(b) shows the relative reflectivity change ($\Delta R/R_{\text{Gold}}$) for different FEL wavelengths as a function of Φ . We find that the switching at shorter wavelengths requires higher fluence as compared to longer wavelengths. For instance, at wavelength = 6.3 μm , the relative reflectivity ($\Delta R/R_{\text{Gold}}$) of 60 % is obtained at $\Phi \sim 10 \text{ mJ}/\text{cm}^2$ but appears to be saturating. Similar saturation is observed for wavelength λ between 8 and 10 μm , but with a much higher $\Delta R/R_{\text{Gold}}$ in the range of 70% to 90 %. For longer wavelength, the switching was found to be more efficient. At a wavelength of 87.4 μm , the $\Delta R/R_{\text{Gold}}$ of reached $\sim 95 \%$ for $\Phi = 2 \text{ mJ}/\text{cm}^2$. The switching fluence Φ_c scales approximately linearly with FEL frequency as depicted in the inset of Fig. 4(b). This contrasts the usual quadratic scaling, which is expected from the relation between the plasma frequency and the charge carrier density.²¹ In order to explain the observed behavior of the photoinduced reflectivity and the scaling of the critical fluence Φ_c , we have performed a numerical simulation of the reflectivity from the

photoexcited surface layer of Ge:Au using the approach described in Ref. 28. It takes into account the inhomogeneous plasma that is photoexcited only within a thin layer near the surface. The thickness of the photoexcited layer is defined by the absorption depth in Ge at 800 nm, which is around 230 nm.²⁹ The calculated results are shown in Fig. 4(c) for the parameters as in the experiment. One can see a good qualitative agreement with the measured data except for the pump fluences Φ , which are higher in the experiment by approximately factor of 3. This can be related to the saturation of optical absorption in Ge,²² or a lower carrier mobility in Ge:Au as compared to the literature values used in the simulation. Remarkably, our simulation reproduces well the linear scaling of Φ_c with the FEL frequency. This demonstrates that it is important to consider the finite sub- μm thickness of the reflecting plasma layer.

Finally, we measured the transmitted FEL power without optical pumping of the Ge:Au switch for different FEL wavelengths (6 to 14 μm). We find excellent transmission properties, with 96 % to 99 % for an average incident power of 2 W. This remarkable negligible absorption of the FEL power in spite of the gold doping and high incident FEL power. At this point, it is important to mention that the actual average intracavity power may reach few hundreds of Watts depending on the FEL wavelength. This is more than two orders of magnitude higher than in the present experiments. However, the focal lengths of the intracavity optics and correspondingly the FEL spot size on the real cavity dumper are expected to be several times larger than those employed in our experimental setup. The FEL energy flux \propto pulse energy/(beam radius)², thus, for example, for an intracavity power of 200 W and 5 times larger spot size the FEL energy flux would be only 4 times more than in our measurements with 2 W of the FEL power. This suggests that for real operations the Ge:Au photoswitch would require somewhat more cooling (possibly cryogenic), which should be technically feasible. Based on our experiments which are close to intracavity conditions we expect the Ge:Au to withstand high loads of intracavity FEL power and to allow a stable FEL operation.

In summary, we have tested Ge:Au wafers as an optical switch under fluences close to what is expected for the real operation of a cavity dumper for the FELBE FELs. The Ge:Au sample exhibits sub-ns recovery times, much shorter than the FEL pulsing period of 77ns. We found a pump fluence of 6 mJ/cm² to be sufficient for pulse picking (at kHz repetition rate) with at least 50 % dumping efficiency for FEL pulses over a broad spectral range from 6 μm to 90 μm . These studies will pave the way for FEL experiments that require higher peak power and

less average power, and is also suitable for investigating materials with longer carrier lifetime (ms range) than the FEL pulsing period.

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DATA AVAILABILITY

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

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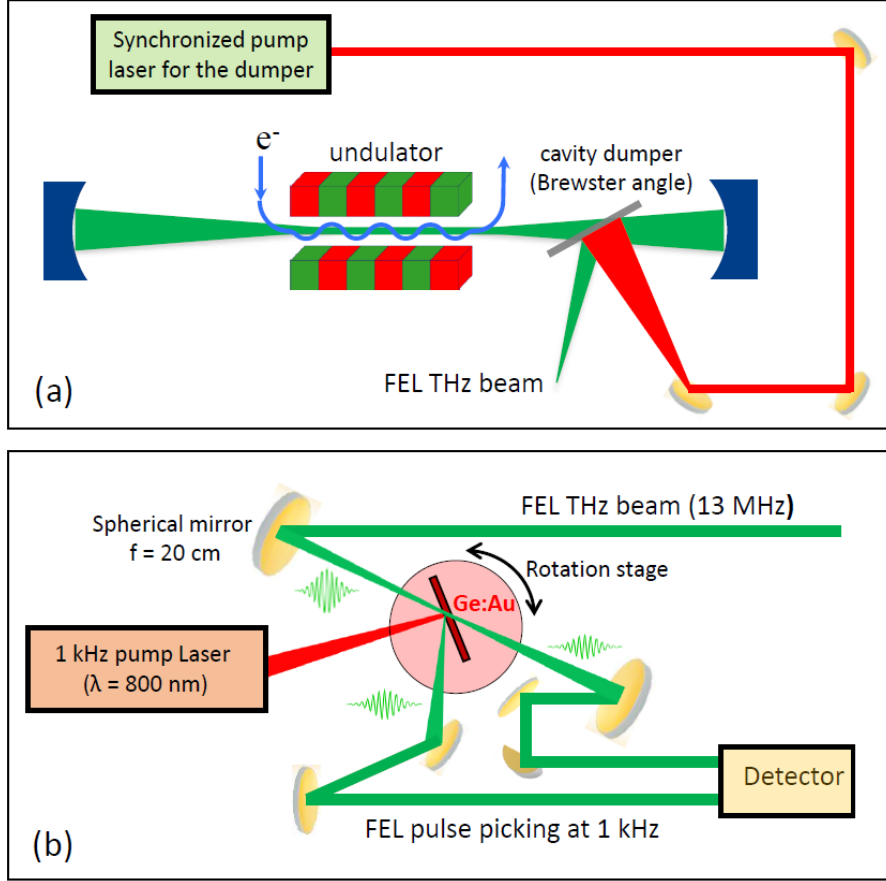


Figure 1. (a) Principle of the cavity dumping using a photoexcited semiconductor switch placed at the Brewster's angle. (b) Experimental setup. The sample is mounted on a thermoelectric cooler and it is at the Brewster's angle with respect to the FEL beam. Both reflected and transmitted pulses can be detected.

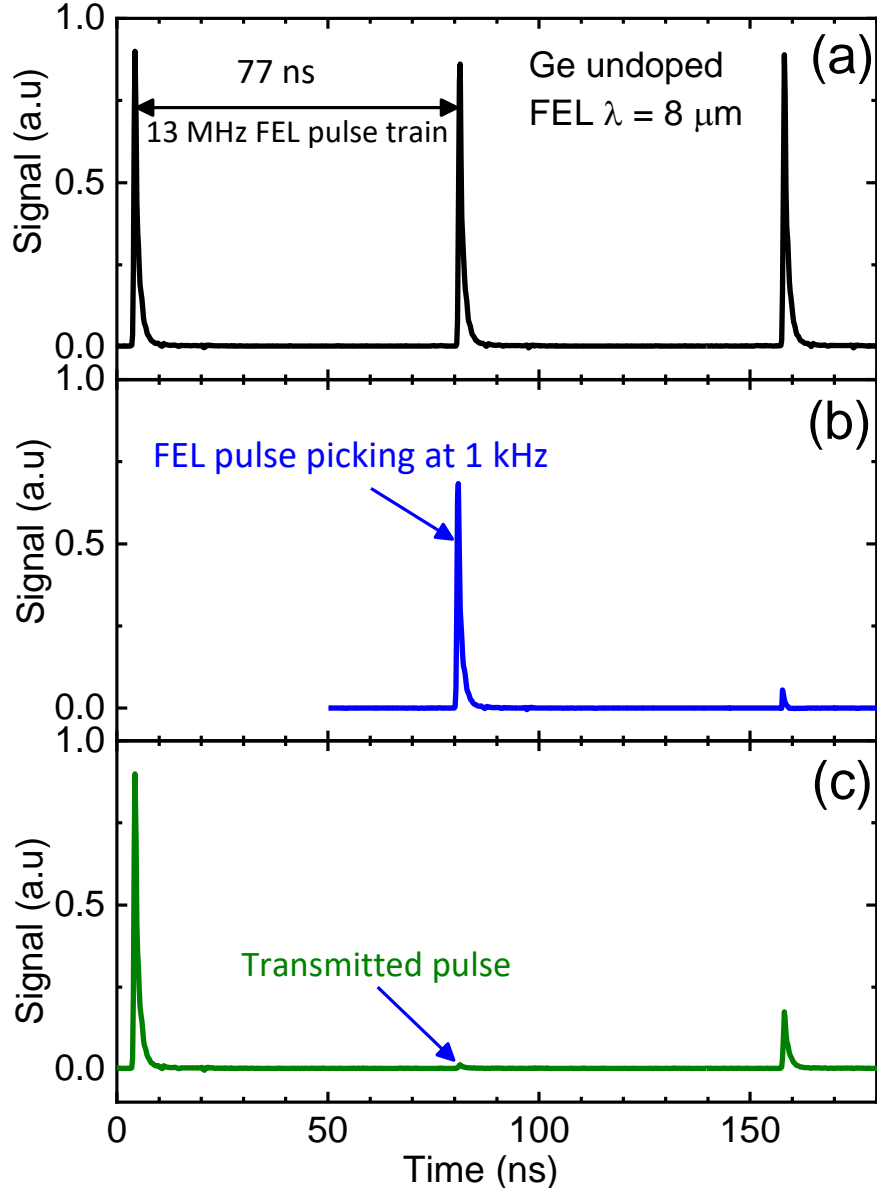


Figure 2. (a) FELBE pulse train with 13 MHz repetition rate, the wavelength is 8 μm . (b) reflected and (c) the transmitted signal of the pure Ge wafer pumped at 1 kHz repetition rate with a pump fluence $\Phi = 10 \text{ mJ/cm}^2$ at the time of 80 ns.

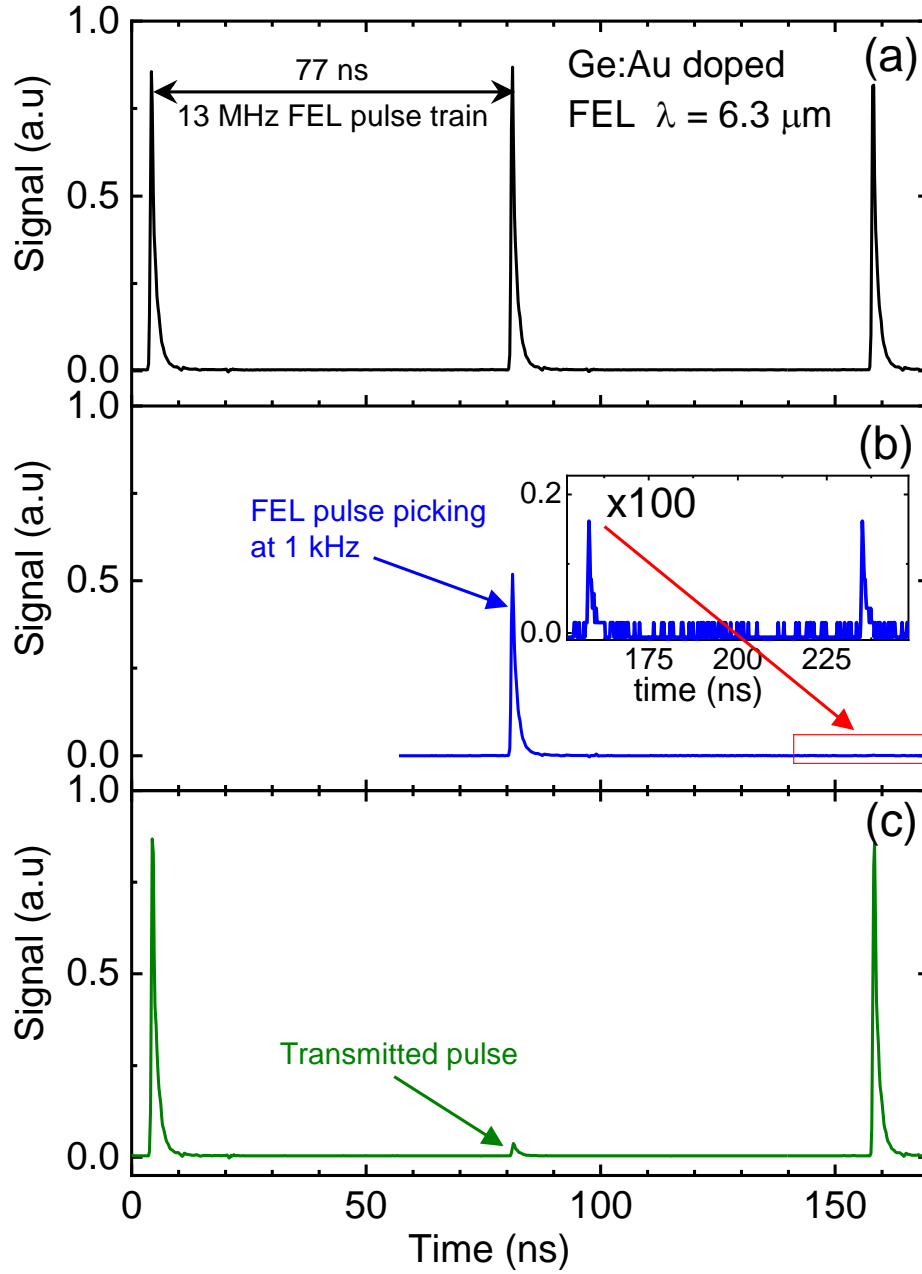


Figure 3. (a) FELBE pulse train with 13 MHz repetition rate, the wavelength (λ) is $6.3 \mu\text{m}$. (b) reflected and (c) the transmitted signal of the doped Ge:Au wafer pumped at 1 kHz repetition rate with a pump fluence $\Phi = 10 \text{ mJ/cm}^2$ at the time of 80 ns. The inset of figure (b) depicts the enlarged signal after the picked pulse.

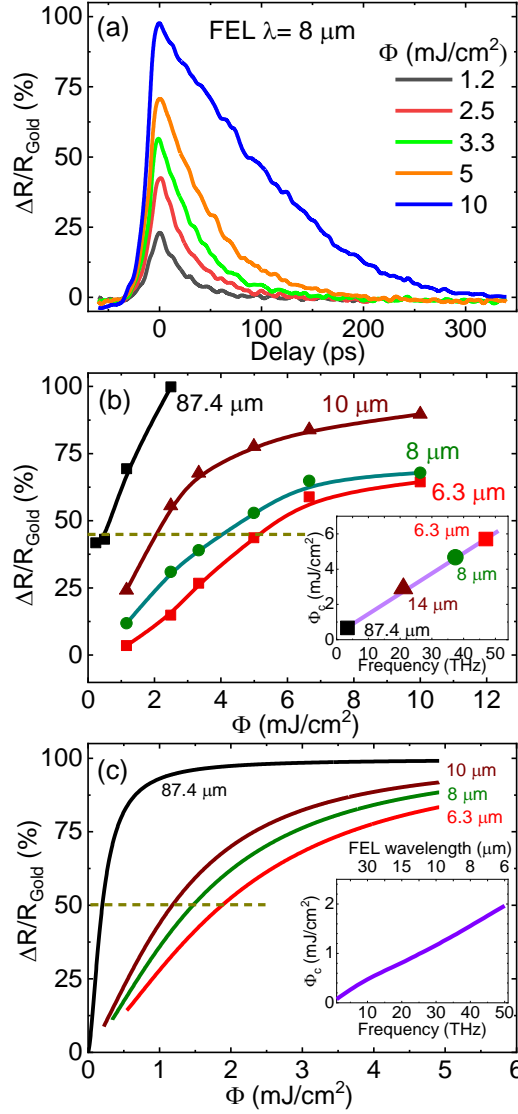


Fig. 4. (a) Photoinduced reflectivity changes ($\Delta R/R_{\text{Gold}}$) for the Ge: Au sample as a function of the time delay between FEL pulses and the pulses from a 1 kHz Ti: Sapphire Amplifier system at a wavelength was 800 nm. (b) The fluence (Φ) dependence of the relative photoinduced reflectivity ($\Delta R/R_{\text{Gold}}$) change for different FEL wavelengths ($\lambda \sim 6.3$ to $87.4 \mu\text{m}$). The inset depicts the fluence (Φ_c) required to couple out FEL frequencies with an efficiency of at least 50 % reflectivity. The violet line is a guide to the eye. (c) The simulated photoinduced reflectivity as a function of the excitation fluence for the same FEL wavelengths. The inset shows the model results for Φ_c as a function of frequency.