threshold for both short (inset) and long delay times. The arrow indicates the lowest reflectivity, representing the highest temperature reached in the process since the reflectivity of bismuth has a negative temperature coefficient [11]. Figure 2(b) shows the lowest reflectivity as a function of fluence. The lowest reflectivity changes almost linearly with the laser fluence. Similar results have also been observed when bismuth was excited above the melting threshold [4].

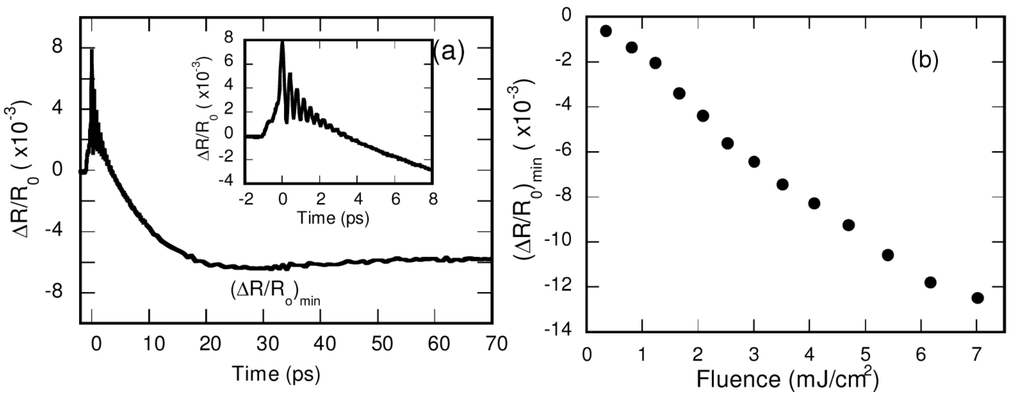
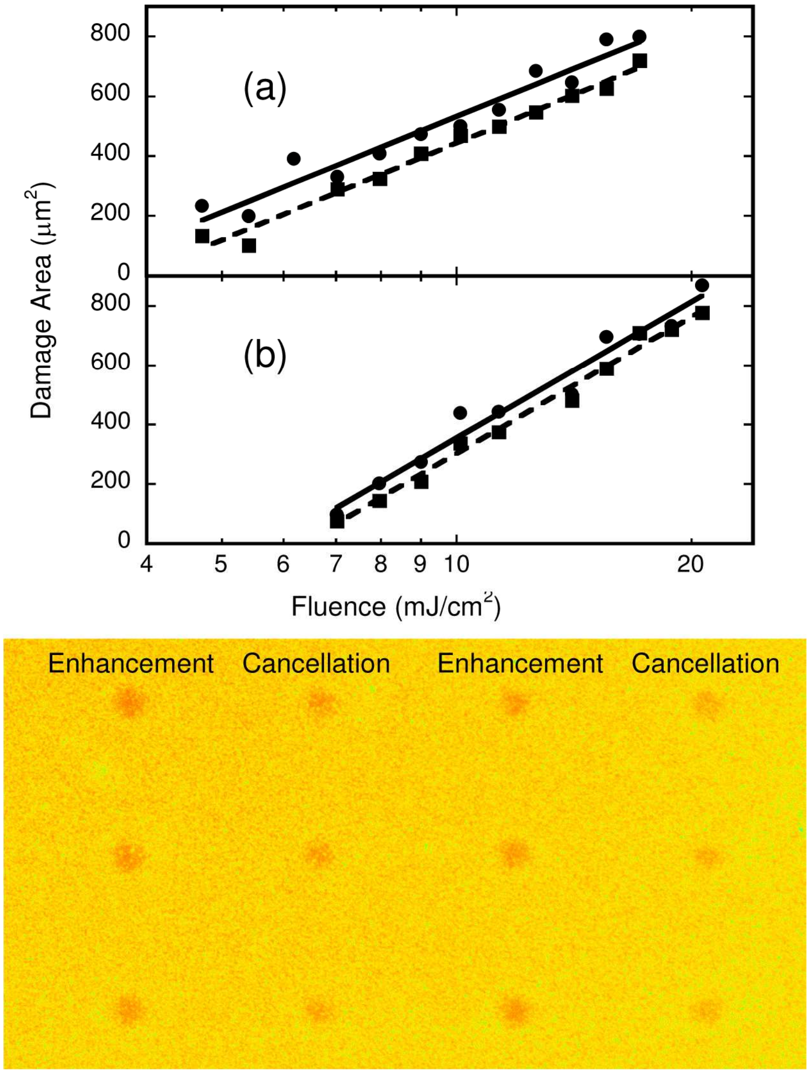


Fig. 2. (a) Pump-probe measurements for fluence of 3.1 mJ/cm^2 for long and (inset) short delay times. The arrow shows the time when the lowest reflectance or highest temperature is obtained. (b) The minimum reflectivity as a function of laser fluence.

The laser fluence required to cause permanent damage using a single 400 nm pulse is about 14 mJ/cm^2 (see below). At laser fluences above the melting threshold and up to the single-shot damage threshold, coherent oscillations can still occur in the first few picoseconds due to the time required to transfer energy from electrons to the lattice. The effect of phonon oscillations at these higher fluences on materials damage was investigated in this work. The damaged areas produced by 5,000 pulses were measured. Using 5,000 pulses instead of single pulse allows determination of the damage areas more accurately. Figure 3 shows the multi-pulse damage area as a function of laser fluence for 5,000 (a) double-pulse and (b) four-pulse excitation designed to enhance and cancel coherent phonon oscillations. It is seen that for both cases, the damage areas caused by enhanced phonon oscillations are larger than those when phonon oscillations are suppressed. An example of surface optical image produced by phonon enhancement and phonon cancellation with a total laser fluence of about 7 mJ/cm^2 is shown in Fig. 3(c). Experiments were repeated in each case (six times each shown in the figure) to ensure reproducibility of the results. The image shows a clear difference in the size of the damaged area between phonon enhancement and phonon cancellation. Also, the damaged area appears darker with phonon enhancement compared with that produced with phonon cancellation.

#132059 - $15.00 USD Received 21 Jul 2010; revised 2 Sep 2010; accepted 2 Sep 2010; published 10 Sep 2010



(c)

Fig. 3. Area of visible damage as a function of the incident pulse fluence for 5,000 (a) double-pulse and (b) four-pulse pulse trains designed to enhance and cancel coherent phonon oscillations. For the double-pulse, the normalized pulse amplitudes were 1.0 and 0.15 with a pulse separation of 406 fs for enhancement and 610 fs for cancellation. For the four pulse measurements the normalized pulse amplitudes were 1.0, 0.15, 1.0 and 0.15. For double-pulse excitation a minimum total fluence of 3.15 mJ/cm^2 and 3.85 mJ/cm^2 for enhancement and cancellation of phonon oscillations, respectively and for four-pulse excitation a minimum total fluence of 5.86 mJ/cm^2 and 6.33 mJ/cm^2 for enhancement and cancellation of phonon oscillations, respectively. (c) Optical image of surface damage produced by double-pulses for phonon enhancement (column 1 and 3) and phonon cancellation (column 2 and 4). The total laser fluence is about 7 mJ/cm^2.

The damage regression method [17] was used to extract the minimum fluence required for damage. Due to the Gaussian intensity profile of the incident beam, the damaged area increases with the natural logarithm of the total pulse fluence. When plotted on a semi-log scale, the minimum fluence required for damage can be extrapolated as the x-intercept of the fitted line. It was found for double-pulse excitation, the minimum total fluence required for damage with 5,000 pulses was 3.2 mJ/cm^2 for phonon enhancement and 3.9 mJ/cm^2 for

#132059 - $15.00 USD Received 21 Jul 2010; revised 2 Sep 2010; accepted 2 Sep 2010; published 10 Sep 2010

phonon cancellation. For four-pulse excitation, a total laser fluence of 5.9 mJ/cm^2 wasneeded to damage for phonon enhancement and 6.3 mJ/cm^2 was needed for phonon cancellation. The difference in the fluence required between the double-pulse and four-pulse excitation can be caused by the differences in the fluences of the individual pulses - the fluences of the individual pulses in the two-pulse pulse train are higher. In addition, the four-pulse excitation allows phonon oscillations to increase over the duration of the pulse train, but does not excite the same maximum phonon amplitude compared with two-pulse excitation as seen in Fig. 1.

The differences in the damage area and the minimum fluence for damage when the coherent phonon oscillations are enhanced and cancelled are indicative of the effect of the coherent phonons on materials phase change. From a phenomenological point of view, amplifying phonon oscillations decreases the stability of the lattice and assists phase change. Results shown in Fig. 3 provide experimental evidence that coherent control of phonon oscillation can affect material's damage.

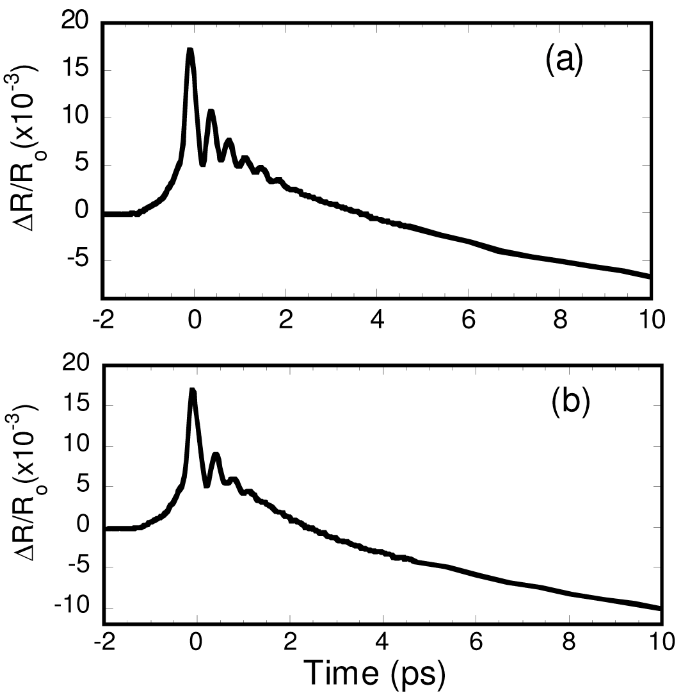


Fig. 4. Pump-probe measurements for fluences of (a) 11.3 mJ/cm^2 and (b) 15.6 mJ/cm^2 at excitation wavelength of 400 nm. Although excited at fluences close to and above the single shot damage threshold, rapidly dephasing oscillations are still visible.

We would like to point out that recent measurements using X-ray and electron diffraction have shown when the single pulse fluence is higher than 23 mJ/cm^2 at 800 nm excitation, "non-thermal' melting occurs at about 200 fs, faster than the coherent phonon oscillation period [18,19]. This fluence corresponds to ~14 mJ/cm^2 at 400 nm. For four-pulse pulse trains used in Fig. 3, the laser fluences of individual pulses are all below 14 mJ/cm^2. For double-pulse pulse trains, the fluence of the first pulse can be around or slightly above 14 mJ/cm^2 for the highest fluences used in Fig. 3 as the ratio of the first to second pulse was about 6:1, yet the phonon oscillations affect the material's damage. Phonon oscillations near and above 14 mJ/cm^2 were observed in our optical measurements. Figure 4 shows measurement results of a single 400 nm pulse excitation with a fluence of (a) 11.3 mJ/cm^2 and (b) 15.6 mJ/cm^2. (Since the laser fluence is close to and exceeds the single shot damage threshold, it was necessary to translate the sample during the measurement.) In our measurements, we found evidences of oscillations beyond 200 fs at fluences where "non-thermal" melting is expected. One possible reason is that in Refs. 18 and 19, the experiments were performed using thinner bismuth films

#132059 - $15.00 USD Received 21 Jul 2010; revised 2 Sep 2010; accepted 2 Sep 2010; published 10 Sep 2010

(30 nm and 50 nm) whereas in our experiments, a 100 nm-thick film was used. The thicker film in our work allows hot carriers to diffuse further away from the surface, therefore lowering the surface temperature and increasing the damage threshold.

4. Conclusions

In conclusion, we show that by using temporally shaped femtosecond pulse trains, we are able to control the coherent phonon oscillations in bismuth. When the pulse-to-pulse frequency of the incident pulses matches the oscillation frequency of the coherent phonons thus enhancing the amplitude of phonon oscillations, there is more damage than when the pulses cancel phonon oscillation. Therefore, controlling the coherent phonon oscillation has a direct effect on the amount of damage in materials such as bismuth.

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