which should be cited to refer to this work.

Ultrafast transient generation of spin-density-wave order in the normal state of BaFe$_2$As$_2$ driven by coherent lattice vibrations

Measurements of the coherent oscillations

1. Spectrally resolved and spectrally integrated measurements of the coherent oscillatory response

We measure amplitude and phase sensitive transient changes of electromagnetic waves in reflection geometry as a function of the delay time t_D between the pump and probe pulses. In the spectrally resolved (two-dimensional) measurement, the entire terahertz (THz) electric field transient is recorded together with its pump-induced change at any given time delay t_D. The time evolution of the induced change in the reflected THz electric field is shown in Fig. S1a. By combining a two-dimensional data set with the equilibrium complex conductivity data obtained by spectroscopic ellipsometry, the dynamics of the spectrally resolved complex THz conductivity is obtained as shown in Figs. 1 and 2. Such studies require extensive data acquisition times of several hours. Therefore, it is practical to study the spectrally integrated response. In a one-dimensional measurement, changes in the electric field at a fixed electro-optic sampling (EOS) delay time t_{EOS} are recorded (Fig. S1b). Here we choose $t_{EOS} \approx 0.01$ ps corresponding to the line marked by the red arrow in Fig. S1a. There the induced change in the electric field caused by the collapse of the spin density wave (SDW) gap is maximum. The spectrally integrated scan also provides an overall relaxation profile although it lacks the information on changes in optical constants. Such one-dimensional measurements were performed to obtain a systematic temperature dependence of the oscillation amplitudes across the SDW transition, an example of which is shown below in Fig. S1b.
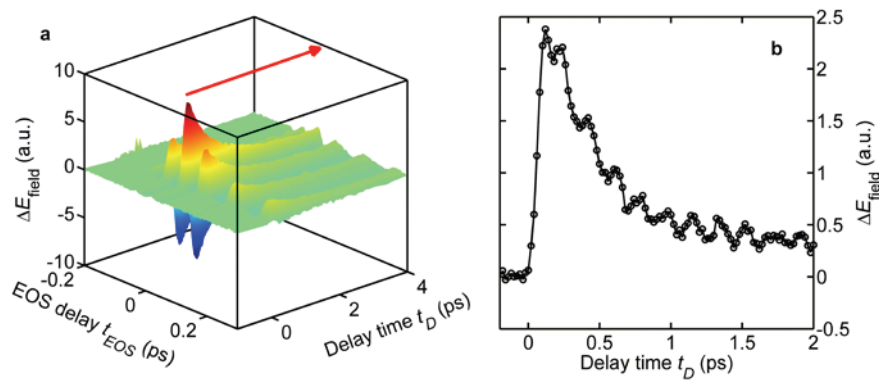
Figure S1 | Raw data of the time domain measurements. a, Pump induced changes in the electric field of the probe pulse. The corresponding time evolution of the THz conductivity is shown in Fig. 1b of the paper. **b,** An example of one-dimensional spectrally integrated response obtained at T_L = 134 K and Φ = 530 μJ/cm^2 which corresponds to the measurement at a fixed EOS delay along the arrow in Fig. S1a.

## 2. Oscillations of the SDWs in the SDW state

The coherent oscillations appear as modulations smaller than 10^{-3} in the amplitude and smaller than 10^{-3} radian in the phase of the reflectivity transient. In the SDW state, the large electronic signal, originating from the collapse and recovery of the SDW gap, is about an order of magnitude stronger than the oscillatory component. Therefore, it is extremely difficult to spectrally resolve the oscillatory response in detail. However, we could still detect the oscillations when spectrally integrating the induced conductivity changes around the SDW induced peak in conductivity as shown in Fig. S2.
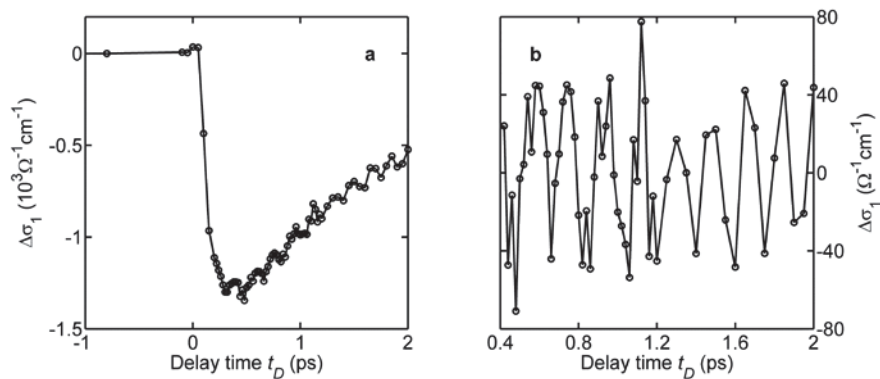
Figure S2 | Oscillations of the SDW-induced peak at T_L = 30 K. a, $\Delta\sigma_1$ averaged over 83-124 meV range recorded at the excitation density of Φ = 710 μJ/cm^2 is shown. **b,** The oscillatory part of the photo induced conductivity change after subtracting the contribution of the dominant electronic response.

## 3. Studies of the coherent phonon dynamics using near-infrared (NIR) pump-NIR probe technique

The NIR pump-NIR probe experiments were performed in reflection geometry employing 60 fs optical pulses (800 nm carrier wavelength) at a repetition rate of 250 kHz. Details of the experimental setup can be found in Ref. [S1]. Figure S3 shows the fast Fourier transformed spectrum of the oscillatory signal obtained in the NIR region together with that in the MIR region. It demonstrates a well-defined phonon mode at 5.46 THz confirming that the coherent oscillations both regions originate from the A_{1g} vibration of arsenic ions.
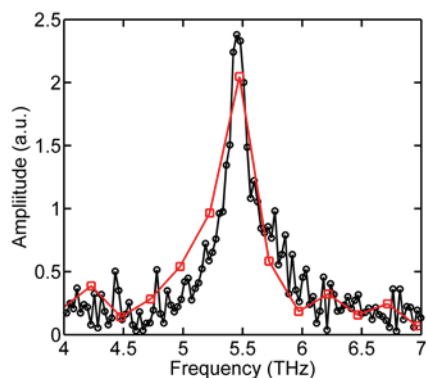
Figure S3 | The Fourier transformed spectra of the oscillatory signal. The black circles are from a measurement at $T_L = 21$ K and $\Phi = 200$ μJ/cm^2 in the NIR region and the red squares are from a one-dimensional measurement at $T_L = 113$ K and $\Phi = 490$ μJ/cm^2 in the MIR region.

4. Determination of amplitudes of the oscillatory component

The temperature dependence of the amplitude of the oscillatory component from the measurements in the infrared region is shown in Fig. 4a. At every temperature, the amplitude was obtained from a fit to the measured oscillatory signal with a fixed mode frequency of 5.46 THz. Since the damping time of the coherent oscillations is substantially longer than the time window, we have used a simple A*sin($\omega t + \phi$) function to extract the amplitude of the oscillatory response. One example is shown in Fig. S4. The oscillation amplitudes in the NIR pump-NIR probe measurements shown in Fig. 4b were obtained from Lorentz oscillator fits to the peak in the Fourier transformed spectra.

4
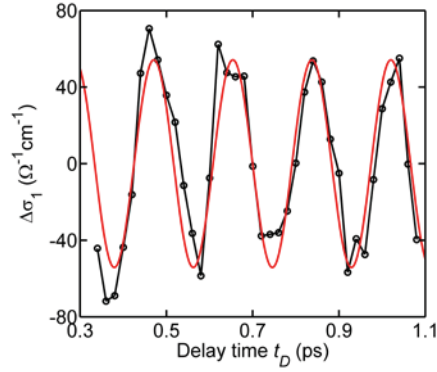
Figure S4 | A fit to the oscillatory signal of the $\Delta\sigma_1$. The oscillatory signal of $\Delta\sigma_1$ is obtained from the average over 41-60 meV range with the electronic (incoherent) response subtracted (black circles). The presented data are measured at T_L = 134 K and at Φ = 530 μJ/cm^2. The red line is fit to a sinusoidal function.

5. Estimate of the amplitude of coherent lattice displacement

An experimental determination of exact values of coherent lattice displacements represents an important challenge for emerging imaging techniques for ultrafast structural dynamics, once the relevant time resolution and sensitivity are achieved. The order of magnitude of the relevant lattice displacement may, however, be estimate already from the NIR reflectivity modulation $\Delta R/R$ and equilibrium dielectric constants based on equations (11) and (12) in Ref. 27:

$$\frac{1}{R}\frac{\partial R}{\partial E_c}Q_0^2 = \frac{\Delta R}{R}\frac{\mathrm{Im}(\varepsilon)}{\hbar\Omega_0^2\rho c}\frac{\Phi}{1-R},$$

Here E_c is the center frequency of the pulse, Ω_0 is the coherent phonon frequency, ρ is the mass density of the oscillating ions in the material, c is the speed of the light, ε the dielectric constant, and Φ is the absorbed fluence. A measured value of $\Delta R/R \approx 2\times10^{-5}$ at Φ = 200 μJ/cm^2 at 134 K results in $Q_0 \approx 0.24$ pm. Since $Q_0 \propto \Delta R \propto \Phi$ (see equations (11)

5

and (12) in Ref. 27), we obtain $Q_0 \approx 0.64$ pm at 530 μJ/cm^2. Taking into account the finite time resolution of our experiment of 70 fs in comparison to the coherent phonon oscillation cycle of 183 fs leads us to an estimate of Q_0 of approximately 0.8 pm.

References:

[S1] Schäfer, H. *et al*. Disentanglement of the electronic and lattice parts of the order parameter in a 1D Charge Density Wave system probed by femtosecond spectroscopy. *Phys. Rev. Lett*. **105**, 066402 (2010).