A Microscopic Description of Displacive Coherent Phonons

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We develop a Hamiltonian-based microscopic description of laser pump induced displacive coherent phonons. The theory captures the feedback of the phonon excitation upon the electronic fluid, which is missing in the state-of-the-art phenomenological formulation. We show that this feedback leads to chirping at short time scales, even if the phonon motion is harmonic. At long times this feedback appears as a finite phase in the oscillatory signal. We apply the theory to BaFe$_2$As$_2$, explain the origin of the phase in the oscillatory signal reported in recent experiments, and we predict that the system will exhibit red-shifted chirping at larger fluence. Our theory also opens the possibility to extract equilibrium information from coherent phonon dynamics.

Introduction.— The development of femtosecond laser pumps has led to new probes of complex metals whereby systems are driven out of equilibrium with the aim to study their relaxation dynamics [1–3]. Simultaneously, pump-probe setups allow the fascinating possibility to study phenomena that have no analog in equilibrium physics, such as the transient excitation of coherent optical phonons [4–7]. A “coherent” phonon is excited when the relevant atoms of the crystalline solid, which are macroscopic in number, vibrate with identical frequency and phase (see Fig. 1(a)). This is to be contrasted with incoherent motion triggered by quantum and thermal fluctuations in equilibrium where, from atom to atom, the frequencies and phases are uncorrelated. More recently it has been recognized that the physics of Floquet excitations can be made experimentally accessible via coherent phonon excitations [8, 9].

Experimentally, a typical signature of a coherent phonon excitation is an oscillatory signal on a decaying background in time-resolved spectroscopic probes such as X-ray spectroscopy, photoemission and reflectivity measurements. Coherent phonons have been studied in a variety of materials that include semiconductors [10–12], semimetals [13–19], transition metals [20], Cu-based [21–24] and the Fe-based [25–33] high temperature superconductors, charge density wave systems [34–38], as well as Mott [39–42] and topological [43–45] insulators.

On the theory side, this phenomenon is usually described either as displacive excitation of coherent phonons (DECP) [36–47] or as impulsive stimulated Raman scattering (ISRS) [48, 49]. In the former mechanism photoexcitation leads to a shift in the equilibrium position of the phonon [40–47], while in the latter the electromagnetic radiation provides a short impulsive force to the atoms [48, 49]. Note, if the photoexcitation does not involve crossing phase boundaries, then typically only the fully symmetric Raman $A_g$ phonon is excited in DECP. It has been argued that in absorbing medium these two mechanisms are not distinct [50]. Using the above concepts, first principles calculations have been successfully applied to understand coherent phonon dynamics in a variety of systems [51–55].

The purpose of this work is to develop, within the conceptual framework of DECP, a microscopic Hamiltonian-based description of coherent phonons in an environment where the timescale for the photoexcited carriers to thermalize is rather short, such as a metal with gapless charge excitations. As we show below, the microscopic formulation provides a better treatment of electron-phonon interaction compared to the phenomenological model that is currently used to analyze experimental data [46]. In particular, our theory captures how the coherent phonon excitation modifies the electronic fluid, and how this modification feeds back on the coherent phonon dynamics.

The main advances of our work compared to the phenomenological theory of Zeiger et al [46] are the following. (i) Including the lattice feedback effect leads to a richer description of the dynamics. In particular, we show that at short time scales this leads to chirping or tempo-
ral variations of the oscillation frequency, while staying within a harmonic description of the coherent phonons. On the other hand, at long times the feedback leads to a finite phase in the oscillatory signal. The origin of this phase is distinct from that in the phenomenological DECP theory\cite{46}, and it is likely to be dominant quantitatively. Importantly, the theory predicts that the sign of the phase is determined by whether the chirping is red or blue shifted. (ii) A Hamiltonian formulation opens the possibility of extracting microscopic equilibrium information from coherent phonon studies. (iii) The microscopic formulation can be refined systematically using methods of many-body to deal with various interaction effects.

**Model & Formalism.**—We consider a multiorbital electronic system interacting with a zero wavevector uniform $A_{1g}$ phonon mode. It is described by the Hamiltonian

$$
\mathcal{H} = \sum_{k,a,b,\sigma} \left[ \epsilon_{(k)ab} - \mu \delta_{ab} \right] c_{k\sigma}^\dagger c_{kb\sigma} + N \hbar \omega_0 (b^\dagger b + 1/2) + \lambda \sum_{k,a,b,\sigma} C(k)_{ab} c_{k\sigma}^\dagger c_{kb\sigma} (b^\dagger + b),
$$

\(\epsilon(k)_{ab}\) describes the dispersion in an orbital basis, and \(\mu\) is the chemical potential. \(c_{k\sigma}\) and \(c_{kb\sigma}\) are electron creation and annihilation operators, respectively, with lattice wavevector \(k\), orbital index \(a\), and spin \(\sigma\). The operators \((b^\dagger, b)\) describe creation and annihilation operators for the $A_{1g}$ phonon with frequency \(\omega_0\), and \(N\) is the total number of sites. Electron-phonon interaction is described by \(\lambda C(k)_{ab}\), where \(\lambda\) is a dimensionless small parameter. For clarity we ignore the phonon modes that are not coherently generated. We also ignore electron-electron and phonon-phonon interaction. Later, we comment on their effects.

After the pump the initial dynamics of the system is dominated by light-matter and by electron-electron interactions. However, as time and angle resolved photoemission (tr-ARPES) experiments have shown\cite{19, 30}, due to electron-electron scattering the electronic subsystem equilibrates after a time \(\tau_r\) of order few tens of femtoseconds. At longer times an instantaneous electronic temperature \(T(t)\) can be defined. In this work we focus on the regime \(t \gg \tau_r\). Accordingly, we assume \(\tau_r \to 0\), such that the effect of the laser pump can be modeled as inducing a temperature quench of the electrons. We assume that the electronic temperature relaxation is characterized by a timescale \(\tau_e\), and is described phenomenologically by

$$
T(t) = T_L + (T_H - T_L) e^{-t/\tau_e},
$$

where \(T_L \equiv T(t = 0^-) = T(t \to \infty)\), and \(T_H \equiv T(t = 0^+)\) [see Fig. 1(b)].

The dimensionless mean atomic displacement \(u \equiv (b^\dagger + b)\) follows the equation of motion

$$
\left( \partial_t^2 + 2\gamma \partial_t + \omega_0^2 \right) u = F(t),
$$

where the out-of-equilibrium force is

$$
F(t) = -\frac{2\omega_0}{N} \lambda \sum_{k,a,b,\sigma} C(k)_{ab} \langle c_{k\sigma}^\dagger (t) c_{kb\sigma} (t) \rangle \mathcal{H}_{T(t)}.
$$

Here \(\langle X \rangle_{H,T(t)} \equiv \text{Tr}[\rho X]/\text{Tr}[\rho]\) and \(\rho \equiv |n\rangle \langle n| e^{-E_n/T(t)}\), where \(|n\rangle\) and \(E_n\) are the eigenfunctions and eigenvalues, respectively, of \(\mathcal{H}\) in Eq. 1.

Our goal is to capture, at least qualitatively, the feedback of the coherent phonon on the electron fluid, for which it is sufficient to evaluate the force to second order in \(\lambda\). At this order \(u(t)\) can be treated as a classical variable fluctuating in time, and \(F(t)\) can be evaluated using linear response theory. We get

$$
F(t)/(2\omega_0) = -(\dot{\mathcal{O}})_{\mathcal{H}_0,T(t)} - \int_{-\infty}^{\infty} dt' \Pi_{T(t)}(t-t') u(t'),
$$

where \(\Pi_{T(t)}(t-t')\) is the response function associated with the weighted electron density operator \(\hat{\rho} \equiv (\lambda/N) \sum_{k,a,b,\sigma} C(k)_{ab} c_{k\sigma}^\dagger c_{kb\sigma}\), and \(\mathcal{H}_0 \equiv \mathcal{H}(\lambda = 0)\). Since all the averages involving electronic operators from now on are defined with respect to \(\mathcal{H}_0\), henceforth we do not mention it explicitly. Note, as discussed in the Supplementary Material (SM)\cite{60}, \(\Pi_{T(t)}(t-t')\) is a function not just of \((t-t')\), but also of \(t\) via its dependence on temperature \(T(t)\). Moreover, the Fourier transform of the response function \(\Pi_{T(t)}(\Omega)\) coincides with the equilibrium retarded phonon self-energy \(\Sigma_{ph}(\Omega)\) evaluated to second order in \(\lambda\) and at temperature \(T\)\cite{60}. At this stage it is also evident that, if needed, effects of electron-electron interaction can be systematically introduced in the evaluation of \(F(t)\).

The fact that the coherent phonon is a well-defined excitation implies that the retardation in \(\Pi_{T(t)}(t-t')\) is weak, and it is sufficient to expand in frequency \(\Pi_{T(t)}(\Omega) \approx \pi(T) + i\Omega \gamma(T)/\omega_0\). Here \(\pi(T) \equiv \Pi_R(\Omega = 0, T)\) and \(\gamma(T)/\omega_0 \equiv \partial^2 \Pi_R(\Omega, T)/\partial T^2\), where \(\Pi_R(\Omega, T)\) are the real and imaginary parts of \(\Pi(\Omega, T)\), respectively. Note, in general, both \(\pi(T)\) and \(\gamma(T)\) are time dependent through their \(T(t)\) dependencies. In the following we simplify the discussion by assuming the decay rate \(\gamma\) is constant, even though the current formulation can handle time-dependent decay rates. This gives

$$
(\partial_t^2 + 2\gamma \partial_t + \omega_0^2) u = f(t),
$$

and

$$
f(t) \equiv -2\omega_0 \left[ \langle \mathcal{O} \rangle_T - \langle \mathcal{O} \rangle_{T_L} + \{\pi(T) - \pi(T_L)\} u(t) \right]
$$

is the instantaneous out of equilibrium force. In the above we added by hand the terms involving \(\langle \mathcal{O} \rangle_{T_L}\) and \(\pi(T_L)\). The former term is a constant that simply sets the zero of the displacement \(u\) to be the atomic position at \(T_L\). The latter term, proportional to \(u\), can be absorbed in the definition of \(\omega_0\). Henceforth, \(\omega_0\) implies the equilibrium phonon frequency at \(T_L\). Note, using Eq. 2 we now get the behavior that is physically expected, namely

$$
f(t = 0^-) = f(t \to \infty) = 0.
$$

The functions \(\langle \mathcal{O} \rangle_T\) and \(\pi(T)\) are well-defined thermodynamic quantities which, in the absence of a phase
transition, are analytic in $T$. Thus, they can be expanded around $T_L$ and, using Eq. (2), they can be expressed as series in powers of $e^{-t/\tau_2}$. In practice, these series can be truncated after the first few terms. We assume that these two series can be modeled as

$$\langle \dot{O} \rangle_T - \langle \dot{O} \rangle_{T_L} \approx -(X_1/2) e^{-t/\tau_1},$$

$$\pi(T) - \pi(T_L) \approx -(X_2/2) e^{-t/\tau_2},$$

where $X_1 \sim \mathcal{O}(\lambda)$, $X_2 \sim \mathcal{O}(\lambda^2)$, $\tau_1, 2 \sim \tau_2$ are four constants defined by the above approximate relations. We get

$$f(t) = \omega_0 \left( X_1 e^{-t/\tau_1} + uX_2 e^{-t/\tau_2} \right),$$

where the second term is the lattice feedback which can be interpreted as the effect of the change in the electron dispersion due to the coherent phonon excitation. Eqs. (8) and (9) summarize the main results of this work.

**Results.**—(i) Evaluating the force $f(t)$ to linear order in $\lambda$ is equivalent to ignoring the lattice feedback by setting $X_2 = 0$ in Eq. (6). In this limit we recover the phenomenological result of Zeiger et al. [46], namely $u(t) = (X_1/\omega_0)[e^{-t/\tau_1} - e^{-t/\tau_2} \cos(\omega_0 t - \phi_0)/\cos\phi_0]$, with the phase $\phi_0 \sim \max[\gamma/\omega_0, 1/(\omega_0 \tau_1)]$. However, the detection of a coherent phonon necessarily implies that in a typical experimental situation

$$\omega_0 \gg \gamma, 1/\tau_{1/2},$$

and so $\phi_0 \ll 1$ is negligible. As we show below, keeping the lattice feedback term also leads to a finite phase, and this latter is quantitatively more significant than $\phi_0$.

(ii) Finite $X_2$ leads to a richer dynamics and a modified solution. Taking $[\gamma/\omega_0, 1/(\omega_0 \tau_{1/2})] \to 0$ we get

$$u(t) = \frac{X_1 e^{-t/\tau_1}}{\omega_0 - X_2 e^{-t/\tau_2}} - \frac{X_1 e^{-t}}{\omega_0 - X_2} \cos[\omega_0 t + \Phi(t)],$$

where

$$\Phi(t) = -\frac{X_2 \tau_2}{2} \left( 1 - e^{-t/\tau_2} \right).$$

Eqs. (8) and (9) summarize the main results of this work.

At face value, the above is a five parameter description of the coherent phonon. However, if the microscopic prescription is followed, $(X_1, X_2, \tau_1, \tau_2)$ can be obtained from the phenomenological parameters $T_H$ and $\tau_2$ defined in Eq. (2) by using the approximate relations of Eq. (5). Furthermore, if the theory to $\mathcal{O}(\lambda^2)$ is quantitatively sufficient, then $\gamma^{-1}$ is the equilibrium phonon lifetime measured by, say, Raman response.

(iii) For $t \lesssim \tau_2$ the feedback $\Phi(t)$ describes temporal variation of the oscillation frequency, i.e., chirping, with a frequency variation $\Delta \omega_0 \sim -X_2/2$, see Fig. 2. On the other hand, for $t \gg \tau_2$ we get a finite residual phase $\phi \equiv \Phi(t \to \infty) = -X_2 \tau_2/2$, see Fig. 2. Note, even if $|\Delta \omega_0|/\omega_0 \ll 1$ and the chirping is not experimentally observable at low fluence, the phase $\phi = (\Delta \omega_0/\omega_0)(\omega_0 \tau_2)$ can be substantial since it involves the large parameter $\omega_0 \tau_2$, c.f., Eq. (7).

(iv) The chirping discussed here is related to the temperature, and hence, time dependence of the phonon frequency due to electron-phonon interaction. This is to be contrasted with other mechanisms of chirping discussed in the literature such as that due to phonon anharmonicity [15] and carrier diffusion [17] [22] [23].

Equilibrium Raman spectroscopy of BaFe$_2$As$_2$ shows that the $A_{1g}$ phonon frequency softens with increasing temperature [56]. Simultaneously, the phonon lifetime [54] has an atypical temperature dependence across the magnetic transition of BaFe$_2$As$_2$ which is very reminiscent of the $T$-dependence of resistivity [57], implying that the phonon temperature dependencies are likely due to interaction with the electrons. Thus, we conclude that $X_2 > 0$, and we predict that the coherent $A_{1g}$ phonon of BaFe$_2$As$_2$ will show red-shifted chirp at sufficiently high fluence.

(v) Since in our theory the frequency shift $\Delta \omega_0$ and the residual phase $\phi$ both depend on $X_2$, an important conclusion is that red-shifted (blue shifted) chirp is accompanied by negative (positive) residual phase.

Note, the above expectation is indeed correct for the $A_{1g}$ coherent phonon of BaFe$_2$As$_2$, which softens with...
FIG. 3: (color online) Quantitative description of the $A_{1g}$ coherent phonon (frequency $\omega_0/(2\pi) = 5.5$ THz) of BaFe$_2$As$_2$, and comparison with experiment [42]. (a) Calculated equilibrium expectation value of the weighted electron density $\langle \hat{O}\rangle_T$. (b) Solid (black) line: The $T$-dependence in (a) is transformed into a time dependence using Eq. (2) for representative values of the phenomenological parameters $(T_H, \tau_e)$. Base temperature $T_L = 140$ K. Dashed (red) line: Fit using Eq. (5), and estimate of $(X_1, \tau_1)$. (c) Solid lines: temporal variation of X-ray form factor calculated using Eq. (8) at different fluences (FL in mJ/cm$^2$). The table gives estimates of $(T_H, \tau_e)$ used in the calculation. The fit uses $\gamma^{-1} = 5$ ps, which is the equilibrium lifetime [59]. Symbols represent data points extracted from Ref. [42].

Increasing temperature, and for which a negative phase $\phi = -0.1\pi$ has been reported [40] [42]. Indeed, for $T_L \sim 150$ K and $T_H \sim 500$ K, an extrapolation of $\omega_0(T)$ reported in Ref. [60] gives $\Delta\omega_0 = -0.4$ THz, and $X_2/\omega_0 \approx 0.01$. Thus, the $-0.1\pi$ phase can be accounted for if $\tau_2 \sim 800$ fs, which is close to the related time scale measured in Ref. [40].

(vi) We apply the theory quantitatively to the coherent $A_{1g}$ phonon of BaFe$_2$As$_2$, and we successfully compare the result with a recent time-resolved X-ray study [42]. The goal is to model the X-ray data starting from a microscopic tight-binding description [58]. We assume the electron-phonon coupling $C(k)_{ab}$ is such that the hopping integrals and the bandwidths are reduced as the As height increases [59], see SM for details of the microscopic model [60]. Determining the out-of-equilibrium force $f(t)$ to $O(\lambda^2)$ involves the computation of $\langle \hat{O}\rangle_T$ which is simple, and the result is shown in Fig. 3(a). From $\langle \hat{O}\rangle_T$ one can obtain the parameters $(X_1, \tau_1)$ at each fluence using Eqs. (2) and (5), provided $(T_H, T_L, \tau_e)$ are known, as shown in Fig. 3(b) for representative values. $T_L$ is the base temperature of pump-probe experiments, which is usually known. For this study the effect of the feedback, i.e., the $O(\lambda^2)$ correction to the force $f(t)$, is rather weak because at these fluences $X_2/\omega_0 \ll 1$ and, simultaneously, the experimental time scale $t \lesssim \tau_2 \sim 800$ fs is short. Consequently, instead of calculating the feedback microscopically, we use the values $X_2/\omega_0 = 0.01$ and $\tau_2 = 800$ fs estimated above, and we assume they do not vary with fluence in the subsequent fits. Thus, fitting the time dependencies of the X-ray form factors, which are proportional to $u(t)$ given by Eq. (5), is equivalent to a two parameter fit of the data at each fluence, where $(T_H, \tau_e)$ are the fit parameters. This microscopic approach is to be contrasted with a purely phenomenological approach where one needs at least four parameters to fit the data at each fluence. In Fig. 3(c) we show that the two-parameter fit is quite reasonable, given the simplicity of the starting model. Note, the above calculation does not include temperature dependencies of the single electron properties arising due to electron-electron interaction. While such interaction effects can be incorporated in the current formalism, it is beyond the scope of the current work.

Conclusions.— We have developed a microscopic theory of displaceable coherent phonons which is more powerful than the standard phenomenological description. Our theory captures the modification of the electronic energy levels due to the phonon excitation, and how this change feeds back on the phonon dynamics. This effect of electron-phonon interaction leads to chirping at short time scales, and at long times it appears as a finite phase in the oscillatory signal. We successfully apply the theory to the $A_{1g}$ coherent phonon of BaFe$_2$As$_2$, explain the origin of the phase in the oscillatory signal reported in recent experiments, and we predict that the system will exhibit red-shifted chirping at larger fluence.

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Supplementary Material for “A Microscopic Description of Displacive Coherent Phonons”

Structure of $\Pi_{T(t)}(t - t')$

The response function used in the main text is defined by

$$\Pi_{T(t)}(t - t') \equiv i\theta(t - t')\langle \hat{\Theta}(t'), \hat{\Theta}(t) \rangle|_{\mathcal{H}_0, T(t)}, \quad (S1)$$

where $\hat{\Theta} \equiv (\lambda/N) \sum_{k,a,b,c} C(k)^{ab|c} \hat{c}_{ka} c_{kb} c_{kc}$ is the weighted electron density operator, $\mathcal{H}_0 \equiv \mathcal{H}(\lambda = 0)$, and the Hamiltonian $\mathcal{H}$ is given by Eq. (1) in the main text. In equilibrium $\Pi_{T(t)}(t - t')$ is a function of $(t - t')$ only, but this is no longer the case out-of-equilibrium. Here, we discuss the $t$ and $t'$ dependencies of $\Pi_{T(t)}(t - t')$. We write the response function in the Lehmann representation where the time structure can be made explicit

$$\Pi_{T(t)}(t - t') = i\theta(t - t') \sum_{n,m} \langle \langle n | \hat{\Theta} | m \rangle \rangle^2 e^{i(t-t')(E_n-E_m)} \left( e^{-\beta(t)E_m} - e^{-\beta(t)E_n} \right), \quad (S2)$$

where $E_n$ and $|n\rangle$ are respectively a complete set of eigenenergies and eigenstates of the Hamiltonian $\mathcal{H}_0$. We see from [S2] that the response function is a function of the time difference $(t - t')$, and that the explicit time $t$ dependence enters only through the electronic temperature $T(t)$. We can then define the Fourier transform of the response function with respect to the time difference $(t - t')$ evaluated at the electronic temperature $T(t)$

$$\Pi_{T(t)}(\Omega) = \sum_{n,m} \langle \langle n | \hat{\Theta} | m \rangle \rangle^2 \frac{1}{E_n - E_m - \Omega + i\eta} \left( e^{-\beta(t)E_n} - e^{-\beta(t)E_m} \right), \quad (S3)$$

where $\eta$ is an arbitrarily small positive constant that ensures the convergence of the Fourier transform. By inspection, we see that the response function in frequency domain [S3] is the equilibrium retarded phonon self-energy evaluated to second order in the electron-phonon interaction ($\lambda^2$) at temperature $T(t) = \beta(t)$.

Solution of the differential equation for $u(t)$

As discussed in the main text, if the instantaneous out-of-equilibrium force $f(t)$ is evaluated to second order in electron-phonon interaction the theory captures the modification of the electronic dispersion due to the coherent phonon excitation, and how that feeds back upon the dynamics of the phonon itself. Taking this feedback into account the differential equation governing the atomic displacement $u(t)$ is given by [see Eqs. (4) and (6) in main text]

$$(\partial_t^2 + 2\gamma\partial_t + \omega_0^2) u = f(t) = \omega_0 \left( X_1 e^{-t/\tau_1} + uX_2 e^{-t/\tau_2} \right). \quad (S4)$$

The parameters $(\omega_0, X_1, X_2, \gamma, \tau_1/2)$ are defined in the main text. Here we discuss the solution of the above differential equation subject to the initial conditions $u(0) = 0$ and $\partial_t u(0) = 0$, and in the experimentally relevant limit of $[\gamma/\omega_0, 1/(\omega_0\tau_{1/2})] \rightarrow 0$. The equation of motion (S4) is linear, the solution is then the sum of the homogeneous and particular solution $u(t) = y_h(t) + y_p(t)$. We first discuss the homogeneous solution, then following the same method we give the particular solution. We start from the following ansatz for the homogeneous solution

$$y_h(t) = \sum_{n=0}^{\infty} a_n e^{kn t} + cc, \quad (S5)$$

with $k_n = i\omega_1 - \gamma - n/\tau_2$, and $\omega_1 = \sqrt{(\omega_0)^2 - \gamma^2}$. We insert (S5) into the homogeneous equation, and obtain an equation for the coefficients $a_n$

$$a_n (k_n^2 + 2\gamma k_n + (\omega_0)^2) = \omega_0 X_2 a_{n-1}, \quad a_0 (k_0^2 + 2\gamma_0 k_0 + (\omega_0)^2) = 0. \quad (S6)$$

Since $k_0$ satisfies the equation $(k_0^2 + 2\gamma_0 k_0 + (\omega_0)^2) = 0$, $a_0$ is then an arbitrary complex constant. We solve the coupled equation (S6), and obtain for $a_n$

$$a_n = a_0 (\omega_0 X_2)^n \prod_{m=1}^{n} \frac{1}{k_m^2 + 2\gamma k_m + \omega_0^2} = \frac{a_0}{n!} \left( \frac{iX_2 \tau_2}{2\omega_1} \right)^n \prod_{m=1}^{n} \frac{1}{1 + (2i)(m/\tau_2 \omega_0)} \approx \frac{a_0}{n!} \left( \frac{iX_2 \tau_2}{2\omega_0} \right)^n, \quad (S7)$$
where in the last step we took the limit \([\gamma/\omega_0, 1/(\omega_0\tau_2)] \to 0\), the homogeneous solution then reads
\[
y_h(t) = a_0 e^{i(\omega_0 - \gamma)t} \sum_n \frac{1}{n!} \left( \frac{iX_2\tau_1}{2\omega_0} e^{-t/\tau_2} \right)^n + cc = a_0 e^{-\gamma t} \left( e^{i\omega_0 t + \frac{X_2\tau_2}{2\omega_0} e^{-t/\tau_2}} \right) + cc, \tag{S8}
\]
We replace \(a_0 = \frac{1}{2} A e^{i\psi}\) and finally obtain for the homogeneous solution
\[
y_h(t) = Ae^{-\gamma t} \cos \left( \omega_0 t + \frac{X_2\tau_2}{2\omega_0} e^{-t/\tau_2} + \psi \right), \tag{S9}
\]
where \((A, \psi)\) are arbitrary constants to be determined from the initial conditions. We follow the same method to find the particular solution, we start from the ansatz
\[
y_p(t) = \sum_{n=0}^{\infty} b_n e^{\alpha_n t}, \tag{S10}
\]
with \(\alpha_n = -1/\tau_1 - n/\tau_2\). We insert \([S10]\) into the equation of motion \([S4]\), and get an equation for the coefficients \(b_n\)
\[
b_n(\alpha_n^2 + 2\gamma_0\alpha_n + (\omega_0)^2) = \omega_0 X_2 a_{n-1}, \quad b_0(\alpha_0^2 + 2\gamma_0\alpha_0 + (\omega_0)^2) = \omega_0 X_1. \tag{S11}
\]
We solve the coupled equations and obtain
\[
b_n = \frac{\omega_0 X_1 (\omega_0 X_2)^n}{(1/\tau_1)^2 - 2\gamma(1/\tau_1) + (\omega_0)^2} \prod_{m=1}^{n} \frac{1}{\alpha_m^2 + 2\gamma\alpha_m + \omega_0^2} \approx \frac{X_1}{\omega_0} \left( \frac{X_2}{\omega_0} \right)^n, \tag{S12}
\]
where in the last step we took the limit \([\gamma/\omega_0, 1/(\omega_0\tau_1/2)] \to 0\), the particular solution then reads
\[
y_p(t) = \frac{X_1}{\omega_0} e^{-t/\tau_1} \sum_n \left( \frac{X_2 e^{-t/\tau_2}}{\omega_0} \right)^n = \frac{X_1}{\omega_0 - X_2 e^{-t/\tau_2}} e^{-t/\tau_1}. \tag{S13}
\]
We use the initial conditions \(u(0) = 0\) and \(\partial_t u(0) = 0\) to calculate the arbitrary constants \((A, \psi)\). The solution in the limit \([\gamma/\omega_0, 1/(\omega_0\tau_1/2)] \to 0\) reads
\[
u(t) = \frac{X_1}{\omega_0 - X_2 e^{-t/\tau_2}} e^{-t/\tau_1} - \frac{X_1}{\omega_0 - X_2} e^{-\gamma t} \cos \left( \omega_0 t + \frac{X_2\tau_2}{2\omega_0} (e^{-t/\tau_2} - 1) \right), \tag{S14}
\]
where we finally recognize Eq (8) of the main text.

**Quantitative description of the A_{1g} coherent phonon in BaFe$_2$As$_2$**

In the main text we discussed a quantitative application of the microscopic theory to describe the A_{1g} coherent phonon in BaFe$_2$As$_2$. The results of the calculations are summarized in Fig. (3) of the main text. In this section we discuss the details of the microscopic calculations.

First, we define the microscopic Hamiltonian used to describe BaFe$_2$As$_2$. In the unfolded Brillouin zone \([S1]\) where the system can be described in an effective 1Fe/unit cell, and in an orbital basis the Hamiltonian is given by
\[
\mathcal{H} = \sum_{k,a,b,\sigma} \left[ \epsilon_{ab}(k) - \mu \delta_{ab} \right] c_{k\alpha\sigma}^\dagger c_{kb\sigma} + N\hbar \omega_0 \left( b^\dagger b + 1/2 \right) + \lambda \sum_{k,a,b,\sigma} C(k)_{ab} c_{k\alpha\sigma}^\dagger c_{kb\sigma}(b^\dagger + b), \tag{S15}
\]
where the electronic kinetic part \(\epsilon_{ab}(k)\) is a tight-binding fit of the LDA band structure taken from Ref. \([S2]\), \(c_{k\alpha\sigma}^\dagger\) creates an electron in orbital \(a\) with momentum \(k\) and spin \(\sigma\), \(C(k)_{ab}\) are the electron-phonon matrix elements, and \(\lambda > 0\) is a tuning parameter of the electron-phonon interaction. The electrons can hop from one Fe site to another via the As site. Therefore, one expects that an increase of the dimensionless arsenic height \(u = (b^\dagger + b)\) is accompanied by a reduction of the hopping-integrals and the bandwidths \([S3]\). Taking into account this physical expectation, we found that a simple way to model the electron-phonon matrix element is to assume
\[
C(k)_{ab} = |t_{mn}|_{ab}(k), \tag{S16}
\]
where \([t_{nn}]_{ab}\) is the diagonal nearest-neighbour part of the Hamiltonian \(H_0\), i.e., the kinetic part \(\epsilon_{ab}(k)\).

Second, we describe the calculation of the out-of-equilibrium force \(F(t)\) [see discussion following Eq. (2) in the main text]. To first order in \(\lambda\) this involves the calculation of the thermal average of the weighted electron density operator. From Eq. (3) of the main text we get

\[
\langle \hat{O} \rangle_{H_0, T} \equiv \frac{\lambda}{N} \sum_{k, a, b, \sigma} C(k)_{ab} \langle \hat{c}_{k a \sigma}^\dagger \hat{c}_{k b \sigma} \rangle_{H_0, T} = \frac{\lambda}{N} \sum_{k, \nu, \sigma} \tilde{C}(k)_{\nu \nu} n_F \left[ \xi_\nu(k) - \mu(T), T \right],
\]

where the last equality is written in the band basis. Here \(n_F\) is the Fermi function, \(\xi_\nu(k)\) is the energy of an electron in the band \(\nu\) with momentum \(k\), \(C(k)_{\nu \nu}\) is the electron-phonon matrix elements in the band basis, and \(\mu(T)\) is the chemical potential at temperature \(T(t)\). We assume that there is no electronic diffusion \([S4]\), and that the particle number is conserved during the pump-probe cycle. We divide the Brillouin zone into a \((10 \times 10 \times 10)\) grid, and diagonalize \(H_0\) at each point of the grid to obtain the electronic dispersion \(\epsilon(k)\). The chemical potential is then calculated by solving the particle number conservation equation numerically. We assume that the tuning parameter \(\lambda \approx 0.25\) and calculate \(\langle \hat{O} \rangle_{H_0, T}\) for temperatures ranging from 0 to 3500(K), see Fig. 3(a) in the main text. This \(T\)-dependence can be transformed into a time dependence using Eq. (2) of the main text provided we have an estimate of the phenomenological parameters \((T_H, \tau_e)\), see the solid (black) line of Fig. 3(b) in the main text. Henceforth, the base temperature is taken as \(T_L = 140\) K. The resulting time dependence can be modeled by a single decaying exponential using Eq. (5), first line, of the main text. This leads to an estimate of \((X_1, \tau_1)\), see dashed (red) line of Fig. 3(b) in the main text.

Next we discuss the evaluation of the \(\lambda^2\) contribution to the force \(F(t)\). In principle, the microscopic theory gives a clear definition of this term and the associated feedback effects, see Eq. (3) of the main text. In practice, evaluating \(\pi(T)\) requires keeping track of the temperature dependence of the electron self-energy due to electron-electron interaction, which is known to be important for the iron based systems. Not only is this beyond the scope of the current work but, as we argue below, for the fluences used in Ref. [S5] the \(\lambda^2\) contribution is rather weak. Consequently, we simply estimate this contribution from the following arguments. We take advantage of the fact that, to \(\lambda^3\) accuracy, \(\pi(T)\) can also be identified as the equilibrium phonon self-energy whose \(T\)-dependence can be inferred from equilibrium Raman measurement of \(\omega_0(T)\) [S6]. For \(T_L = 140\) K and \(T_H \sim 500\) K, an extrapolation of \(\omega_0(T)\) reported in Ref. [S6] gives \(\Delta\omega_0 = 0.4\) THz, and therefore \(\Delta\omega_0 / \nu \approx 0.01\), see Eq. (9) of the main text. This small fraction implies that the \(\lambda^2\) contribution to the force \(F(t)\) is unimportant. Furthermore, if we assume that the experimentally reported phase \(\phi = -0.1\pi\) [S5] [S7] can be identified with \(\Phi(t \to \infty) = -X_2 \tau_2 / 2\) (see Eq. (9) of the main text), we get \(\tau_2 \approx 800\) fs. In the following we do not consider the \(\pi(T)\) dependence of \((X_2, \tau_2)\). Note, implicitly we assume that the phonon self-energy is dominated by electronic contribution. Phenomenologically, this is supported by the fact the phonon lifetime reported in Ref. [S6] has an atypical temperature dependence across the magnetic transition of BaFe2As2 which is very reminiscent of the \(T\)-dependence of resistivity [S8].

Thus, from the above considerations, for a given fluence we are able to quantify \(u(t)\) given by Eq. (8) of the main text, provided we have an estimate of \((T_H, \tau_e)\).

Finally, we compare our calculated arsenic displacement \(u(t)\) with that measured in time resolved X-ray scattering [S5] for a fluence range of 0.7 to 3.5 (mJ/cm\(^2\)). The intensity is convolved with a Gaussian pulse to account for the limited time resolution [S5]. The arsenic displacement \(u(t)\) is fitted using Eq. (8) of the main text. In the kinematic approximation [S5], the variation of the intensity is proportional to the arsenic displacement and is given by

\[
\frac{\Delta I}{I_0}(t) = \frac{C}{\tau_r \sqrt{\pi}} \int_0^\infty e^{-\left(\frac{t-t_r}{\tau_r}\right)^2} X_1 \left( e^{-\gamma t_r} - e^{-\gamma t} \cos \left[ \omega_0 t + \Phi(\tau) \right] \right) dt ,
\]

where \(C\) is a proportionality constant independent of fluence, \(I_0\) is the equilibrium intensity, \(\Delta I\) is the variation of intensity out of equilibrium, \(\tau_r \approx 96\) fs is the experimental resolution of the probe-pulse, \(f = \frac{\omega_0}{2\pi} = 5.5\) THz is the frequency of the coherent mode, \(\Phi(t) = -\frac{X_2 \tau_2}{2} (1 - e^{-t/\tau_2})\), and \([X_1, \tau_1, X_2, \tau_2, \gamma]\) are defined in the main text. Note that for time \(t \lesssim \tau_r\) the quality of the fit is marginally affected by including the feedback \(\Phi(t)\) term. For the estimation of the proportionality constant \(C\) we use the measurements of an independent time resolved ARPES experiment [S7] where the electronic temperature increases from \(T_L = 100\) K to \(T_H = 500\) K for a fluence \(F_L = 0.47\) mJ/cm\(^2\). Assuming that \((T_H - T_L)\) is proportional to the fluence, we expect \(T_H \approx 760\) K for a fluence \(F_L = 0.7\) mJ/cm\(^2\), which gives an estimate of \(C \approx 19.5\) by fitting Eq. (S18) to the X-ray data. Finally for the remaining fluences we estimate \([T_H, \tau_e]\) that gives the best fit between the data of Ref. [S5] (solid symbols in Fig. 3(c) of main text) and the calculated one from Eq. (S18) (lines in Fig. 3(c) of main text).
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