Coherent topological defect dynamics and collective modes in superconductors and electronic crystals

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Abstract
The control of condensed matter systems out of equilibrium by laser pulses allows us to investigate the system trajectories through symmetry-breaking phase transitions. Thus the evolution of both collective modes and single-particle excitations can be followed through diverse phase transitions with femtosecond resolution. Here we present experimental observations of the order parameter trajectory in the normal $\rightarrow$ superconductor transition and charge density wave ordering transitions. Of particular interest is the coherent evolution of topological defects forming during the transition via the Kibble–Zurek mechanism, which appears to be measurable in optical pump–probe experiments. Experiments on CDW systems reveal some new phenomena, such as coherent oscillations of the order parameter, the creation and emission of dispersive amplitude modes upon the annihilation of topological defects, and mixing with weakly coupled finite frequency (massive) bosons.

(Some figures may appear in colour only in the online journal)

1. Introduction

The idea that non-equilibrium phenomena in natural systems can be described in terms of a temporal evolution of a nonlinear Landau expansion of the free energy has lead to theories with as wide a scope as elementary particles to cosmology. In condensed matter systems, where the idea originates, the opportunities for the study of the behaviour of nonlinear systems abound. However, typically, the study of these systems has been limited to near-equilibrium situations, where the system evolves slowly through the transition. Yet both particle physics experiments and cosmology distinctly deal with the temporal evolution of highly non-equilibrium systems. Typically one detects the decay products well after the decay itself, in the aftermath of the symmetry-breaking transition (SBT). Collisions of elementary particles, the subsequent creation of a high-symmetry intermediate state and its decay via symmetry-breaking interactions takes place on timescales which are well beyond the resolution of our current technology. Cosmological experiments being out of reach, experimental cosmological studies of the aftermath of the Big Bang reveal very complex behaviour in its aftermath, still beyond current theoretical understanding. As a result we have little insight into the non-equilibrium conditions close to the critical time $t_c$ of the SBT when the creation of the new state is taking place.

In condensed matter systems the intrinsic dynamics of elementary excitations occur on timescales which are becoming accessible with current femtosecond laser technology. Particularly the normal-to-superconducting state transition and charge density wave transition are of fundamental interest, as examples with different symmetry properties. Thus, typical single-particle (quasiparticle) relaxation times in high-temperature superconductors are on timescales of $10^{-12}$ s [1], while electronic energy relaxation occurs on timescales of $10^{-12}$–$10^{-13}$ s [2]. Many-body collective states have similar characteristic timescales. The so-called Ginzburg–Landau (GL) time $\tau_{GL}$ [3], which enters into the
the characteristic timescale of the collective mode from single-particle excitations. On the other hand, the characteristic timescale of the collective mode in charge density wave systems is typically around \(2\pi/\omega_{\text{AM}} \simeq 0.5 \times 10^{-12} \text{s} \ [4, 5]\). Laser pulses can currently be created with sub-femtosecond duration, so ample resolution is available for the study of the coherent evolution of collective modes in superconducting and charge density wave (CDW) systems. Moreover, by studying the concurrent evolution of the single-particle spectrum, it may be possible to investigate the transient state of the underlying vacuum.

Following the suggestion by Zurek [6] of laboratory experiments to test Kibble’s cosmological model [7], ‘system quench’ experiments were performed in a number of different systems, including superconductors [8, 9], Bose–Einstein (BE) condensates within a trapped atomic gas [10] and polariton BE condensates [11]. Experiments on the Kibble–Zurek (KZ) mechanism so far have concentrated on the statistical analysis of topological defects left behind by the SBT, on timescales long compared to the intrinsic GL time \(\tau_{\text{GL}}\). Quench rates in these systems were relatively slow, the fastest being around \(10^8 \text{K s}^{-1} \ [9]\).

In standard optical pump–probe (P-p) techniques (including THz probe) the modulation of the dielectric constant (reflectivity or absorbance), which can have a number of contributions in electronic systems such as superconductors and CDWs which are in one way or another related to the order parameter \(\psi\): (1) the response due to hot-carrier energy relaxation, (2) the quasiparticle (QP) recombination across the SC or CDW gap, (3) QP recombination across a pseudo-gap, which is often present in these systems, and (4) coherent phonon oscillations due to displacive excitation or impulsive Raman excitation. The coherent phonon oscillations cannot be distinguished from the oscillations of the order parameter with P-p spectroscopy. Moreover, different phonon modes are coupled to \(\psi\) to various degrees, and the dephasing is different for each mode, thus adding further to the complexity of the response from which it is difficult to extract the order parameter. We show that using a three-pulse technique, these responses can be isolated to some extent, allowing us to study the coherent evolution of the order parameter with time through an SBT with very high temporal resolution. The method allows us to study the interactions of the collective mode of the CDW with other modes of different symmetry as well as the annihilation of topological defects, revealing new finite frequency dispersive Higgs-like field excitations released as a result of domain wall annihilation.

2. Quench experiments with a multiple pulse all-optical technique

The principle behind the technique involves the use of multiple pulse trains to control the system and measure the time–response of the order parameter. A schematic diagram of the pulse sequence is shown in figure 1. A strong laser destruction (D) pulse—whose intensity is carefully adjusted to cause a perturbation of appropriate magnitude—is used to rapidly transfer the system from an ordered (broken symmetry) state to the disordered (high-symmetry) state, whereafter the system reverts back to equilibrium through the SBT. The state of the system at any given time after the D pulse is determined by a standard pump–probe spectroscopy sequence, which involves first exciting the system by an additional weak perturbing P pulse delayed by \(\Delta t_{1-2}\) with respect to the D pulse, and subsequently measuring the resulting change of reflectivity \(\Delta R/R\) of the sample by a weak probe (p) pulse (see figure 1). In section 3.1 we will show how the optical response \(\Delta R/R\) can be related to the order parameter in superconductors and CDW systems.

On the level of field theory, the trajectory of the system through the SBT can be modelled within time-dependent Ginzburg–Landau theory with a free energy functional corresponding to the symmetry of the problem in hand. For a charge density wave system, to capture the salient physics, we can assume an order parameter \(\Psi = A(t, r)e^{i\phi}\) and make the assumption that the relaxation on phase \(\phi\) is slow compared to \(\tau_{\text{GL}}\), which means that the trajectory is dominated by the dynamics of \(A(t, r)\) [12, 13]:

\[
\frac{\partial^2 A}{\partial t^2} + \Gamma \omega_{\text{AM}} \frac{\partial A}{\partial t} - \alpha(t, r)\omega_{\text{AM}}^2 A + \omega_{\text{AM}}^2 A^3 - \xi^2 \omega_{\text{AM}}^2 \frac{\partial^2 A}{\partial x^2} = 0, \tag{1}
\]

where \(\omega_{\text{AM}}\) is the angular frequency of the collective amplitude mode, and \(\xi\) is the coherence length. \(\Gamma = \Delta \omega_{\text{AM}}/\omega_{\text{AM}}\) is the damping of the AM, where \(\alpha(t, r)\) is the transient state of the underlying vacuum.
temporally and spatially dependent and is derived from the
usual GL temperature $\alpha(t, r) = 1 - \mu(t, r)$ describing the
control parameter. $\mu(t, r)$ may be $T(t, r)/T_c$, as given by
original GL theory, or may be approximated by an exponential
function which signifies the cooling of the system. The spatial
variation of the light intensity is accounted for by an excitation
function $\mu(t, z) = T_{\text{AM}}(t)/T_c \exp(-z/\lambda)$, where $\lambda$ is the optical
penetration depth. Using experimental values for $\nu_{\text{AM}} = \omega_{\text{AM}}/2\pi = 2.4$ THz, the line-width $\Delta\nu_{\text{AM}} = 0.05$ THz and
cohere length $\xi = 1$ nm [12] we can compute $A(t, z)$.

In three-pulse experiments, the $P$ pulse presents an
additional perturbation which can modify the trajectory,
especially if it occurs close to the critical time $t_c$. In figure 2
we show calculated trajectories of $\psi$ for three different delay
 times $\Delta t_{1-2}$ of the $P$ pulse with respect to the $D$ pulse. The
‘butterfly effect’ is very evident: $\psi$ may end up as either $+1$
or $-1$, critically depending on $\Delta t_{1-2}$.

The equation of motion for a superconductor within the
time-dependent Ginzburg–Landau (TDGL) model is more
complicated because the excitations are charged and $\psi$
is coupled to the electro-magnetic field, and are usually
considered to be overdamped, so they do not include any
second-order time-derivative:

$$u \left( \frac{\partial \psi}{\partial t} + i \Phi \psi \right) = -\alpha r(t, r) \psi - \psi |\psi|^2 - (i \nabla + a)^2 \psi + \eta$$

(2)

$$\nabla^2 \Phi = -\nabla \left[ \frac{i}{2} (\psi^* \nabla \psi - \psi \nabla \psi^*) + a |\psi|^2 \right].$$

(3)

These equations have been used for analytic and numeric
analyses of the dynamics of topological defects (vortices and
phase slips) in superconducting wires, as reviewed in [14, 15].
The vector potential $a$ is written in units of $\frac{\hbar}{e \tau_0}$ ($\Phi_0$ is the
flux quantum, $\xi$ is the coherence length) and the electrostatic
potential $\Phi$ in units of $\frac{e}{\sigma e^2}$ with $e$ being the elementary
charge, $\tau_0$ is the transverse relaxation time and $\hbar$ the reduced
Planck constant. The only dimensionless parameter left in the
equation is the ratio $u \equiv \frac{\tau}{\tau_0}$ between the longitudinal
relaxation time and the transverse relaxation time. Typi-
cally, for high-temperature superconductors $\tau_0 \approx 1$ ps and
$u \approx 5$ [15]. The Langevin noise term $\eta$ introduces microscopic
fluctuations into the dynamics [15].

3. Control of the quench rate and optical response
function

In the simplest approximation, suggested by the usual $\alpha$
parameter in the GL equations, the excitation function $\mu$
can be the system temperature. In the case where the
 electronic system is being discussed, such as superconductors
and CDWs, it is the electronic temperature $T_e(t)$ which is
the relevant control parameter. In recent years, significant
progress has been made in understanding the time-evolution
of electron and lattice temperatures in superconductors and
CDWs following excitation by a laser pulse. The quench
rate is determined by the energy relaxation rate of electrons
at early times up to $\sim 1$ ps, and the lattice $T_l(t)$ on
longer timescales (tens of picoseconds and beyond). The
time-evolution of $T_e(t)$ and $T_l(t)$ respectively, which govern
the quench process, may be estimated using a two-temperature
model [16]:

$$\gamma_e T_e \frac{dT_e}{dt} = -\gamma_e (T_e - T_l) + E(t)$$

and

$$C_l(T) \frac{dT_l}{dt} = \gamma_L(T_e - T_l),$$

where $E(t)$ is a function (Gaussian) describing the energy
density per unit time supplied by the laser pulse. We used the temperature-dependent thermal constants $\gamma_e(T)$
and $C_l(T)$ from experimental data [19] and the measured
energy loss rate $\gamma_e \approx 340$ K ps$^{-1}$ for La$_1.9$Sr$_{0.1}$CuO$_4$ [2].
The dependence of $T_e(t)$ and $T_l(t)$ on the energy density
per pulse $U \equiv \int E(t) dt$ is shown in figure 3(a) for the first
few picoseconds. The red contour lines show when either
$T_e$ or $T_l$ crosses $T_c$. Initially, the rate of cooling $\gamma_e \approx \frac{dT_e}{dt}$
is very rapid until $T_e$ reaches $T_l$. Thereafter, $T_e$ follows $T_l$,
and is dominated by phonon anharmonic decay or phonon
escape and thermal diffusion processes, all of which are much
slower than the electronic energy loss. Thus, to change the
quench rate we can adjust the energy density $U_D$ of the $D$
pulses, so that the system either cools rapidly through electron thermalization, or more slowly by phonon decay and diffusion processes. As shown in figure 3(a), for low $U_D$, only $T'$ exceeds $T_c$, so the quench back through $T_c$ will be fast and dominated by electronic cooling. For larger $U_D$, $T_1$ exceeds $T_c$, so on cooling the quench rate though $T_c$ is significantly slower and dominated by lattice cooling.

3.1. The response function

The key issue is detection of the order parameter through the SBT. Here we show how for superconductors and CDW systems, the response measured in three-pulse experiments is related to the order parameter. While a full kinetic theory which would take into account single-particle and phonon relaxation processes through the transition is beyond reach at present, we can obtain good approximations for the quasiparticle response functions in superconductors and collective mode in CDW systems, respectively, based on well-accepted phenomenological theory.

The temperature dependence of the difference of the optical reflectivity between the superconducting state $R_s$ and the normal state $R_n$ in a superconductor has been found to be described very well by an expression derived from the Mattis–Bardeen formula [20]:

$$R_s(T) - R_n = A \left| \Delta(T) \right|^2 \ln \left( \frac{1.47 \omega}{\Delta(T)} \right)$$

where $\Delta(T) = 1 - (T/T_c)^2$ is usually used to describe the temperature dependence of the gap, $\omega$ is the frequency of light in units of $h$, $R_n$ is the reflectivity in the normal state. We need to calculate the transient change of reflectivity $\delta R$, as the system is evolving through the transition in time, so we explicitly replace $\Delta(T)$ with $\Delta(t)$. Using the 2-fluid model, we substitute $\Delta^2 = \Delta_0^2 n_s = \Delta_0^2 (1 - n_q)$, where $n_s$ is the superfluid density and $n_q$ is the quasiparticle density, and $\Delta_0$ is the gap at $T = 0$. We can relate $\delta R$ to the photo-excited carrier density $n_p$ using the fact that $\delta n_q = n_p$:

$$\delta R = \frac{\partial R}{\partial \delta n_q} = \Delta_0^2 \frac{\partial \Delta_0}{\partial \Delta} \frac{\Delta}{\Delta(t)} n_p.$$  (5)

If we ignore the derivative of the logarithmic correction with respect to $\Delta$, substituting $\Delta_0$ for $\Delta(t)$ in the logarithm, we obtain $R_s(t) - R_n \simeq A \Delta_0^2 \frac{\partial \Delta}{\partial \Delta} \ln \left[ \frac{1.47 \omega}{\Delta_0} \right]$. $\delta R$ then simplifies to:

$$\delta R = \frac{\partial R}{\partial \delta n_q} \delta n_q = \frac{2 \Delta(0)}{\omega^2} \delta \Delta \propto -\frac{\Delta_0^2}{\omega^2} n_p.$$  (6)

Under near-bottleneck conditions, when the QPs and the phonons are in near-equilibrium, $n_p$ is given by [21]:

$$n_p \propto \frac{1/(\Delta(T(t)) + T(t)/2}{1 + B \sqrt{T(t)/\Delta(T(t))} \exp \left[ -(\Delta(T(t)/T(t)) \right]}.$$  (7)

where $\Delta$ is the superconducting gap and $B \simeq v/N(0)h \Omega_c$, where $v$ is the effective number of phonon modes of frequency $h \Omega_c$ per unit cell participating in the relaxation process. Here we have explicitly written the temperature $T$ to be time dependent. $B$ can be determined by fitting the temperature dependence of $\delta R/R$ and $N(0)$ is the density of electronic states at the Fermi energy. Substituting $\Delta_0 \psi(t)$ for $\Delta(t)$ from the solution of equation (3), we obtain:

$$\frac{\delta R}{R} \propto \frac{\Delta_0^2}{2 \omega^2} \left( 1 - 2 \ln \left[ \frac{1.47 \omega}{\Delta(t)} \right] \right)$$

$$\times \frac{1/(\Delta_0 [\psi(t)] + T(t)/2}{1 + B \sqrt{T(t)/\Delta_0} [\psi(t)] \exp \left[ -(\Delta_0 [\psi(t)/T(t)] \right]}.$$  (8)

The response functions using the appropriate constants for La$_{1.5}$Sr$_{0.5}$CuO$_4$ are plotted in figure 3, together with the time-evolution of $T_c(t)$ for $E = 1.6 J \, cm^{-2}$, and the order parameter $|\psi(t)|$ and $|\psi(t)|^2$ for the case of a homogeneous superconductor calculated using equation (2) in the absence of field $a = 0$, and $\Phi = 0$. The experimental reflectivity response $\delta R/R$ is close to the square of the order parameter $|\psi|^2$ over
a large range of $t$, particularly for large $\psi$. The calculated case for an inhomogeneous superconductor which takes into account the depth profile of the laser beam is discussed in section 4, where it is compared with experimental data.

In CDWs, when discussing the transient response of collective amplitude mode, the response is related to the displacement $\Delta r$ of the atoms from the equilibrium position $r_0$, where $\Delta r \propto \psi - \psi_0$, where $\psi_0$ is the equilibrium value of the order parameter. To obtain the optical response, we can expand the dielectric constant near the CDW phase transition in powers of the order parameter $[17]$

$$
\epsilon = \epsilon_0 + \epsilon_2 |\psi|^2.
$$

Here $\epsilon_0$ is the dielectric constant of the high-temperature symmetric phase and $\epsilon_2$ is a real constant.

The relevant response in $D$–$P$–$p$ experiments can be expressed in terms of the difference between the response with and without the $P$ pulse, proportional to $\Delta |\psi|^2$, so to first order $[12]$

$$
\frac{\Delta R}{R} \simeq \frac{\Delta \epsilon}{\epsilon} \propto \psi_{DP}^2(t, r, \Delta t_{12}) - \psi_{D}^2(t, r).
$$

This response has been tested for the case of the CDW transition in TbTe$_3$ $[12]$. We see that by careful design of the experimental probe and the use of three-pulse techniques, it is possible to identify and even isolate the dominant contribution to the optical response which is related to the order parameter, thus providing valuable information of the temporal evolution of $\psi$ by measurement of either single-particle and collective excitations through the SBT.

4. Superconductors: probing vortex dynamics on the picosecond timescale

In figure 4(a) we show the time-evolution of $\Delta R/R$ in La$_{1.9}$Sr$_{0.1}$CuO$_4$ measured with a $P$–$p$ sequence at a sample base temperature of 4 K after a 50 fs laser pulse. All laser pulses have a wavelength of 800 nm. The $D$ pulse energy $E_D = 0.8$ J cm$^{-3}$ is adjusted to heat the sample above $T_c$. (The threshold values for the destruction of the SC state were previously reported in $[20]$ (see footnote 4).)

According to equation (7), the QP amplitude in $P$–$p$ experiments is related to $|\psi|$, which allows us to compare the data with the calculated trajectory based on a solution of equations (2) and (3). The numerical solution calculated without the fluctuation term $\eta$ is shown by the solid line in figure 4(a). Comparing with the measured data, we see that the prediction is remarkably good overall, except for a depression of the order parameter of approximately 10–20% in the region around 10 ps, shown by the dashed line in figure 4(b). Qualitatively, we see that the presence of vortices may explain the depression of the order parameter around 10 ps in La$_{1.9}$Sr$_{0.1}$CuO$_4$. Considering the simplicity of both the TDGL model and the crudeness of the approximations for the response function (equation (7)), the agreement between data and theory is quite reasonable.
Reflectivity was obtained from the Fourier transform of the transient spectrum after the temporal evolution of the order parameter in a related transition have been reported in TbTe$_3$ single-particle and collective excitations though the CDW transition metal tri-tellurides such as DyTe$_3$, which undergoes a transition to a CDW state at 305 K. The temporal evolution of the collective mode spectrum after the D pulse is shown in figure 5. The spectrum was obtained from the Fourier transform of the transient reflectivity $\Delta R/R$ recorded at each $\Delta t_{1-2}$ as a function of $\Delta t_{2-3}$. We identify two modes in the spectra at $\sim$1.68 and $\sim$2.2 THz at long times $\Delta t_{1-2} > 10$ ps. The latter is assigned to the amplitude mode (AM) of the CDW based on its distinct temperature dependence [5], while the former is a phonon mode (PM). At $t \approx 1$ ps, the AM exhibits mixing with the PM. Oscillations of the order parameter are clearly visible at short times and appear as modulations of the intensity of both modes occurring up to $\sim$8 ps. At longer times, the intensity oscillations and the frequency recovers to 2.2 THz after $4 \times 10^8$ ps. No further relaxation takes place beyond this timescale.

In figure 6(a) we show the calculated $\psi(z,t)$ from equation (1) using parameters for DyTe$_3$: $\omega_{AM} = 2.2$ THz, the AM line-width $\Gamma = 0.3$ THz, and coherence length $\xi = 1.3$ nm from [23]. For simplicity, the driving term $\alpha(t,z)$ in equation (1) is assumed to be exponential in time and in space: $\alpha = \exp(-t/\tau - z/\lambda)$, where the penetration depth is given by the experimental value $\lambda = 20$ nm [24]. The calculation predicts the formation of three domains created within the first picosecond, parallel to the surface, with domain walls at $\sim$30 and $\sim$45 nm. After $5$ ps, the domains walls are annihilated, leaving behind a single domain. The event is accompanied by the emission of a finite frequency dispersive amplitude wave travelling towards the surface and into the sample. The velocity of the amplitude wave is seen from figure 6 to be approximately $v_4 \approx 10$ nm ps$^{-1}$. The wave reaches the surface within 6–8 ps, causing a disturbance of the AM, which is visible as a temporal distortion of the AM spectral line-shape. (The inset to figure 5 highlights the distortions of $\omega - \Delta t_{12}$ spectra.) The calculated spectrum (using equation (10)) is shown in figure (b). The predicted distortions have a great deal of similarity with the experimentally observed evolution of the spectrum shown in figure 5. Unfortunately the presence of the phonon interference in this material complicates the detailed comparison between theory and experiment. Similar, although more pronounced distortions were observed in TbTe$_3$, where the interference from other phonons is less problematic [12].

In [25] it is further shown that incoherent intrinsic topological defect dynamics in the related system TbTe$_3$ occurs on a timescale of $\sim$30 ps, similar to the timescale of vortex annihilation dynamics inferred from the experiments on La$_1\,\text{Sr}_0\,\text{CuO}_4$. The experiments show that pinned-defect-related annihilation appears to be present on much longer timescales of $10^{-10} - 10^{-6}$ s.

5. Topological defects in CDWs: coherent dynamics

Transition metal tri-tellurides such as DyTe$_3$ are excellent systems for femtosecond measurements of system trajectories through SBTs to a CDW state. Recently, the evolution of single-particle and collective excitations though the CDW transition have been reported in TbTe$_3$ [12]. Here we show the temporal evolution of the order parameter in a related material, DyTe$_3$, which undergoes a transition to a CDW state at 305 K. The temporal evolution of the collective mode spectrum after the D pulse is shown in figure 5. The spectrum was obtained from the Fourier transform of the transient reflectivity $\Delta R/R$ recorded at each $\Delta t_{1-2}$ as a function of $\Delta t_{2-3}$. We identify two modes in the spectra at $\sim$1.68 and $\sim$2.2 THz at long times $\Delta t_{1-2} > 10$ ps. The latter is assigned to the amplitude mode (AM) of the CDW based on its distinct temperature dependence [5], while the former is a phonon mode (PM). At $t \approx 1$ ps, the AM exhibits mixing with the PM. Oscillations of the order parameter are clearly visible at short times and appear as modulations of the intensity of both modes occurring up to $\sim$8 ps. At longer times, the intensity oscillations and the frequency recovers to 2.2 THz after $4 \times 10^8$ ps. No further relaxation takes place beyond this timescale.
6. Discussion

We have demonstrated that femtosecond optical experiments with multiple pulse techniques open the way to detailed studies of the temporal dynamics of systems undergoing SBTs and can be interpreted in the context of a phenomenological field theory without resort to microscopic theory. The basic analogy with cosmology and elementary particle collisions comes from the fact that the SBT take place in temporally evolving systems with high-temperature initial conditions. Single-particle fermionic and collective bosonic excitations can be unambiguously identified and related to the temporal evolution of the order parameter through the transition, and topological defects (domain walls and vortices) are shown to lead to experimentally observable phenomena. It appears that CDWs are eminently more suitable for the investigation of topological defects than superconductors because the collective mode conveys significantly more information of the system trajectory than single-particle excitations, particularly relating to coherent defect dynamics. We have shown that by adjusting the laser excitation energy density, the quench rate can be varied and controlled, leading to the possibility of investigating domain wall recombination. Further studies as a function of quench rate and temperature may be expected to reveal systematic behaviour, which can be related to the predictions of the KZ mechanism for the generation of topological defects. The evolution of the single-particle response as a function of time after destruction by a laser pulse in the La$_{1.9}$Sr$_{0.1}$CuO$_4$ superconductor suggests that a depression of the order parameter compared with the theory prediction can be understood if vortices are included within an inhomogeneous model, suggesting intrinsic vortex creation and annihilation in La$_{1.9}$Sr$_{0.1}$CuO$_4$ takes place on a timescale of $10–20$ ps. Although one might argue that the discrepancy between the trajectory of $|\psi|$ and the measured response may be attributed to the breakdown of the approximations made in deriving the response function equation (7), these problems are not expected to be important in the region around $10–20$ ps, but rather at shorter times, closer to $t_c$: when $\psi \to 1$, the gap is fully developed, and the QP response is expected to be described well by the Rothwarf–Taylor model in the bottleneck regime [22], where the gap $\Delta(t) \to 1$ and is almost constant, so equation (7) is reasonably valid, as shown in figure 3.

We point out that in the context of more general field theories of related time-evolving systems, apart from the AM, two other bosons observed in CDW systems are of interest. The first is created as a result of the annihilation of domain walls shown in figure 6. Upon annihilation, two finite frequency (massive) $\psi$-waves are emitted which propagate perpendicular to the surface into the sample and towards the surface. Calculations of the ‘wave effect’ on the temporal evolution of the AM spectral shape appear to be confirmed by experiments in DyTe$_3$ (discussed here) and in TbTe$_3$ [12].
The second bosonic excitation of interest is the phonon mode (PM) with an equilibrium frequency of $\omega_{PM} = 1.68$ THz. The time-evolution of this mode is very different from the evolution of the AM: it does not show critical behaviour near $t_c$. Instead its frequency is just slightly renormalized in the low-symmetry state. Before behaviour near $t_c$, the PM frequency is $\omega_{PM} = 1.85 \pm 0.02$ THz. Because its frequency $\omega_{PM}$ in the ordered state is lower than $\omega_{AM} = 2.2$ THz, it crosses the AM as the latter hardens after $t_c$, as indicated in figure 5. At $\Delta t_{12} \simeq 1.3$ ps the AM interferes with the PM, and the phonon frequency is renormalized for $\Delta t_{12} > 2$ ps, to $\omega_{AM} = 1.68 \pm 0.02$ THz.

In experiments where the control parameter is temperature $T$ rather than time, this behaviour is well understood [5]: below the critical temperature $T_c$, as the AM hardens with decreasing $T$, it crosses the PM at some intermediate temperature, displaying mode mixing. The coupling of the AM and the PM in compounds such as DyTe$_2$ is relatively weak, the off-diagonal matrix element being $\delta \approx 0.1 \pm 0.01$ THz [5]. The PMs form a large reservoir of excitations which are weakly coupled to the order parameter. The symmetry of the PM plays an important role in the temporal behaviour. By definition, the order parameter is totally symmetric, (A representation) so even-symmetry modes are expected to couple strongly to it. When the system has inversion symmetry, such as here, (DyTe$_2$ has $D_{2h}$ point group symmetry) the odd-symmetry modes do not couple to the $\psi$ to first order. Odd-symmetry modes thus display some characteristics of weakly interacting massive particles in cosmology. The observed PM has a large mass (frequency) and is weakly coupled to the order parameter, either because it has inappropriate symmetry (in which case the weak coupling comes from higher order interactions), or the matrix element coupling to A symmetry excitations is small. In contrast to the AM and the fermionic excitations which emerge after the SBT takes place, the PM excitations exist before the SBT (i.e. before and after the Big Bang within the cosmological paradigm). By analogy with the PM, dark matter excitations may be thought of as remnant excitations from before $t_c$. As such, they are external to the GL (or Standard) model.

We conclude that building on the analogy between a laser-induced $e-h$ plasma in condensed matter systems and the plasma created in the collision of elementary particles, or with primordial conditions in the Universe, gives us a practical laboratory-scale playground for the exploration of temporally evolving systems though symmetry-breaking transitions. Remarkably, the CDW model system gives rise to two kinds of cosmological analogies, namely of the AM as the analogue to the Higgs boson and odd-symmetry phonons as dark matter excitations. The CDW systems reveal some hitherto unexplored bosonic excitations created under non-equilibrium conditions which may be expected to have observable counterparts in other temporally evolving systems. The reflectivity response function derived for a superconductor allows the investigation of the trajectory of the order parameter through the transition, opening the way to studies of intrinsic vortex dynamics.

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