Mechanisms of nonthermal destruction of the superconducting state and melting of the charge-density-wave state by femtosecond laser pulses

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The processes leading to nonthermal condensate vaporization and charge-density wave (CDW) melting with femtosecond laser pulses is systematically investigated in different materials. We find that vaporization is relatively slow ($\tau_v \sim 1$ ps) and inefficient in superconductors, exhibiting a strong systematic dependence of the vaporization energy $U_v$ on $T_c$. In contrast, melting of CDW order proceeds rapidly ($\tau_m = 50 \sim 200$ fs) and more efficiently. A quantitative model describing the observed systematic behavior in superconductors is proposed based on a phonon-mediated quasiparticle (QP) bottleneck mechanism. In contrast, Fermi surface disruption by hot QPs is proposed to be responsible for CDW state melting.

Photoinduced phase transitions on the sub-picosecond timescale in superconductors and other electronically ordered systems have attracted increasing attention in recent years, partly because of the fundamental desire to control collective states of matter with femtosecond laser pulses, partly because of potential applications in ultrafast phase-change memories, and partly because they may reveal some fundamental insight into the mechanism of high-temperature superconductivity. However, the details of how absorbed photons cause a change of state on ultrafast timescales have so far not been investigated in detail. Recently, a study on La$_2$−$\delta$Sr$_\delta$CuO$_4$ (LSCO) single crystals by Kusar et al. showed that the energy required to vaporise the superconducting (SC) condensate $U_v$ can be determined with reasonable accuracy. These and other measurements on cuprate superconductors since then give values of $U_v$ which are sometimes significantly larger than the experimental condensation energy $U_c$, or the BCS theoretical value, opening the question of mechanism for the destruction of the condensate, the dependence of $U_v$ on doping and on the critical temperature $T_c$. It is also not clear if this occasional large discrepancy between $U_v$ and $U_c$ is peculiar feature of cuprate superconductors, is a consequence of a large gap in the density of states, or depends on the detailed mechanism responsible for the formation of the low-temperature ordered state.

To try and answer these questions, we present the first systematic study of the dependence of $U_v$ on doping and $T_c$ for cuprates and compare our data with the iron pnictides (Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$ and SmFeAsO$_{1-y}$F$_y$), the conventional superconductor NbN and two large-gapped charge-density wave systems K$_{0.3}$MoO$_3$ and TbTe$_3$. We made a special effort to determine the geometrical factors and optical constants as accurately as possible in order to achieve the best possible accuracy in determining $U_v$. We find that the observed systematic behavior reveals the mechanism for breaking up the collective state in high-$T_c$ superconductors to be relatively inefficient and phonon dominated. This is in stark contrast to CDW systems, where the electronic destruction mechanism is faster, more direct and efficient. We present a model to describe the observed systematics in the cuprates and discuss the markedly different mechanisms for the photonic destruction of the low-temperature ordered state in the two types of system.

Our experiments were performed using a standard pump-probe technique with a 800 nm laser with 50 fs pulses as described in ref. A probe wavelength of 800 nm is chosen because it is a well-understood probe wavelength for investigating the superconducting (SC) and pseudogap (PG) response in pump-probe experiments and the best signal/noise ratio as determined by previous studies. The 2-component response, PG and SC, including lifetimes, $T$-dependence, anisotropy and doping dependence have been previously studied at 800 nm and other pump/probe wavelengths as well, including pump/probing in the THz region. The results of these measurements are quantitatively self-consistent, and have all been systematically discussed. The second reason for using 800 nm (1.5 eV) is that the optical constants are known more accurately at this wavelength better than any other, which improves the accuracy of the determination of the vaporization energy. The reason for not using far infrared (FIR) wavelengths in the gap region is that even with transform-limited pulses, the pulses become too long to be able to distinctly detect the SC component and distinguish it from the PG. Hence THz or FIR pulses in the gap region are not suitable for our purpose.

Recently the electron distribution function thermalization has been measured to be ~ 60 fs in LaSCO and ~ 100 fs in YBCO, while the quasiparticle (QP) signal used for detection of the presence of the superconducting state in the cuprates has a characteristic lifetime nearly two orders of magnitude longer and can thus be easily distinguished from the PG and energy relaxation which are both much faster. In order to accurately determine the deposited energy, pump and probe laser beam diameters were accurately measured with calibrated pinholes. The light penetration depth $\lambda_{op}$ and reflectivity $R$, are obtained from published optical conductivity and dielectric function data in each case.
were performed on freshly cleaved YBa$_2$Cu$_3$O$_{7-\delta}$ single crystals with $T_c$ of 90, 63 and 60 K, respectively and Ca$_2$Y$_{1-x}$Ba$_2$Cu$_3$O$_{7-\delta}$ with $x = 0.132$ and $\delta = 0.072$ and $T_c = 73$ K. The photoinduced reflectivity transient $\Delta R/R$ for YBa$_2$Cu$_3$O$_{6.5}$ well below $T_c$ (4 K) and above $T_c$ (at 68 K) are shown in Fig. 1a), while the net superconducting signal, obtained by subtracting the response at $T = 68$ K from the response at $T = 4$ K : $\Delta R_{\text{sc}}/R = \Delta R/R|_{T<T_c} - \Delta R/R|_{T>T_c}$ is plotted in Fig. 1b) for different laser pump pulse fluences. This behavior is generic, so we have shown the raw data only for one sample. $\Delta R_{\text{sc}}/R$ shows a saturation plateau above $F \gtrsim 100 \mu J/cm^2$ which is a signature of the destruction of the SC state.

The normalized magnitude of $\Delta R_{\text{sc}}/R$ for different materials and different doping levels as a function of $F$ is shown in Fig. 2. At low fluence $\Delta R_{\text{sc}}/R$ is linearly dependent on $F$ up to a certain threshold fluence $F$, whereupon it either becomes saturated or continues increasing, but with a different slope. To determine the vaporization threshold fluence $F_T$ from the data in Fig. 2, we use the inhomogeneous excitation model\textsuperscript{[3]} To take into account carrier-density dependent change of $\Delta R/R$ arising from $e-e$ asymmetry in the band structure, we have also included a linear term in the fit to the data in Fig. 2, such that $\Delta R/\Delta R_{\text{sc}} = \Delta R_{\text{sc}}/\Delta R_{\text{sc}} + \phi(F)$. Empirically, we find that in LSCO, $\phi \approx \theta$ while in YBCO samples $\phi(F) \approx \phi_0F$, where $\phi_0$ is a constant. Note that this term has no effect on the values of $F_T$ obtained from the fits to the data in Fig. 2. (As a check, $F_T$ can also be directly read out as the point where $\Delta R_{\text{sc}}(F)/R$ departs from linearity).

A plot of the vaporisation energy $U_v = (1-R)F_T/\lambda_{\text{op}}$ obtained for cuprates, the iron pnictides and NbN is shown in Fig. 3. We observe systematic increase of $U_v$ with $T_c$, giving an approximate square power law dependence $U_v = \eta T_c^3$, with $\eta \approx 1.5 \times 10^{-3}$K$^3$atom$^{-1}$ irrespective of whether the material is overdoped, optimally doped or underdoped.

In Table 1 we compare $U_v$ with the condensation energy $U_c^{\text{exp}}$ obtained from specific heat measurements. In the cuprates, we notice a significant discrepancy between the magnitudes of $U_v$ and $U_c^{\text{exp}}$. While $U_c^{\text{exp}}$ shows a distinct asymmetry between overdoped and underdoped materials with the same $T_c$\textsuperscript{[4]} $U_v$ depends systematically on $T_c$ irrespective of doping level. Thus $U_v$ and $U_c^{\text{exp}}$ show fundamentally different systematics.

To understand this apparent discrepancy between the condensation and evaporation energy, let us now consider the mechanism of energy transfer between photoexcited (PE) carriers and the condensate. After absorption of a photon, the PE electrons and holes lose energy very rapidly through “avalanche” scattering with phonons on a timescale of $\tau_v < 100$ fs for YBCO and $< 60$ fs for LSCO\textsuperscript{[5]} creating a large non-equilibrium phonon population in the process. The phonons whose energy exceeds the gap $\hbar \omega > 2\Delta$ can subsequently excite QPs from the condensate, but these recombine again and so a bottleneck occurs, in which high-frequency phonons are temporarily in quasi-equilibrium with QPs as described by Kusar et al. for LaSrCuO\textsuperscript{[6]} The process eventually causes the destruction of the condensate within $0.5 \sim 1$ ps\textsuperscript{[1]}

The phonons with $\hbar \omega > 2\Delta$ can contribute to the breakup of the condensate, but the low frequency phonons with $\hbar \omega < 2\Delta$ also created in the initial avalanche do not have enough energy to excite QPs, so do not contribute to the evaporation process. This lost energy to low-energy phonons is:

$$U_{\text{lost}} = \int_0^{2\Delta} \Delta f(\omega) D(\omega) \hbar \omega d\omega$$

where $D(\omega)$ is the phonon density of states and $\Delta f(\omega) = f_{\text{NE}}(\omega) - f_{\text{E}}(\omega)$ is the difference between the non-equilibrium and equilibrium phonon distribution functions. $\Delta f(\omega)$ is material-dependent and depends on the
The data for $U_c$ are calculated per metal atom of Cu (planar), Fe, Nb, Tb or Mo. For LSCO and YBCO, the data for $U_c$ are maximum experimentally measured values$^{12}$, $\delta$ in YBCO was changed by anodising in flowing oxygen, while $T_c$s were determined from magnetization measurements. For TbTe$_3$ and K$_{0.3}$MoO$_3$, the electronic part of $U_c$ of the condensation energy is estimated using available values of $\Delta$ and $N_0$.$^{12}$ For YBa$_2$Cu$_3$O$_{7-\delta}$, the penetration depths are$^{12}$ $66\pm15$ nm ($\delta = 0$), $85\pm15$ nm ($\delta = 0.4$), $90\pm15$ nm ($\delta = 0.5$), and $75\pm15$ nm for CaYBCO.

Table I. $T_c$, $F_T$, $U_v$ and $U_c$ for different materials. $U_c$ and $U_v$ are calculated per metal atom of Cu (planar), Fe, Nb, Tb or Mo. $T_c$s are determined from magnetization measurements. For TbTe$_3$ and K$_{0.3}$MoO$_3$, the electronic part of $U_c$ of the condensation energy is estimated using available values of $\Delta$ and $N_0$.$^{12}$ For YBa$_2$Cu$_3$O$_{7-\delta}$, the penetration depths are$^{12}$ $66\pm15$ nm ($\delta = 0$), $85\pm15$ nm ($\delta = 0.4$), $90\pm15$ nm ($\delta = 0.5$), and $75\pm15$ nm for CaYBCO.

![Diagram](image-url) Figure 3. (Color online). $\Delta U_v$ expressed in K per planar Cu as a function of $T_c^2$ for the cuprates. The data for NbN, SmFeAsO$_{0.8}$F$_{0.2}$ and Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$ are included for comparison (in K/Fe or K/Nb). The solid curve is a plot of $U_{\text{lost}}$ using $D(\omega_{ph})$ appropriate from Eq. (2) for YBaCuO (red) and the iron oxy-pnictide (blue). The dashed line is a square law $U_v = \frac{\hbar}{2\Delta}$. The insert shows the phonons with $\omega \approx 2\Delta$ (shaded) which can break pairs. The measured phonon density of states $D(\omega_{ph})$ for YBCO is approximately by a parabola (line).

The total vapourisation energy is the sum of the condensation energy and the lost energy, $U_v = U_c + U_{\text{lost}}$. Since for large gap systems $U_v \gg U_{\text{exp}}$ we have $U_v \simeq U_{\text{lost}}$, so $U_{\text{exp}}$ is thus only a small contribution to $U_v$, explaining why the anomalous doping dependence of $U_{\text{exp}}$ in the cuprates is not displayed by $U_v$.

Comparing $U_v$ for NbN measured using the same technique (Table I), $U_v/U_{\text{exp}} = 1.7$ is considerably smaller than in the cuprates, which can now be understood in terms of Eq. (1) and the insert to Fig. 3. When $2\Delta < \hbar \omega_{\text{Debye}}$ almost all phonons can excite QPs, so $U_{\text{lost}} \rightarrow 0$, and $U_v \simeq U_c$, in agreement with the data. For small-gap superconductors, the optical method can be
used to estimate the superconducting condensation energy. The data on the two iron pnictides listed in Table I also seem to follow the predicted behaviour in Fig. 3.

We now turn our attention to the data obtained using the same technique on two different charge-density-wave materials TbTe$_3$ and K$_{0.3}$MoO$_3$. Both have CDW gaps which are much larger than any phonon energy: $2\Delta_{CDW} \gg \hbar \omega_D$ ($2\Delta_{CDW} \approx$ 125 meV and 75 meV respectively)\[12\]. K$_{0.3}$MoO$_3$ is considered a prototypical Peierls system with a full gap in the electronic density of states opening at the metal-insulator transition\[12\]. Moreover, in KMoO$_3$, the electron-phonon coupling constant ($\lambda_{e-p} \approx 0.35$) is comparable with the cuprates ($\lambda_{e-p} \approx 0.25$ for YBaCuO and $\lambda_{e-p} = 0.5$ for LaSCO respectively)\[13\], so with a phonon-mediated CDW melting mechanism we would expect a very large $U_m/U_c$ ratio. Instead, it is significantly smaller than in the large-gapped cuprates, with $U_m/U_{exp} \approx 3.5$. In TbTe$_3$, the ordered state opens a partial gap only along certain directions on the Fermi surface, the rest remaining ungapped\[12\]. Thus, if the same phonon-mediated vapourisation mechanism were operative, the ungapped CDW system should behave very differently to KMoO$_3$ on one hand, and the superconductors on the other. Comparing $U_c$ with the electronic contribution to the CDW condensation energy\[10\], \[19\], $U_c = N_0 \Delta^2 (1 + \log \frac{2\Delta}{\hbar \omega_D})$, where $E_F$ is the Fermi energy\[10\], we see in Table I that the predicted ratios are $U_m/U_c \simeq 1.3$ for TbTe$_3$ and $U_m/U_c \simeq 1.1$ for K$_{0.3}$MoO$_3$, confirming that $U_{lost} \to 0$ and implying that phonons are not significantly involved in the CDW gap destruction and CDW melting process. Thus, in spite of similar $e-p$ interaction strength and very large gaps, $U_m/U_c$ is consistently smaller in the CDWs than $U_c/U_c$ in cuprates, highlighting the very different mechanism for the destruction of the low-temperature CDW state.

The reason is fundamental: Unlike in superconductors, PE carriers in CDW systems can cause a direct rapid disturbance of the Fermi surface\[12\] on a timescale shorter than the QP recombination time. It is known experimentally\[23\] and theoretically\[24\] that the CDW state is extremely sensitive to charge imbalance, which disrupts the Fermi surface (FS) nesting. In photoexcitation experiments, overall charge neutrality is preserved, but immediately after photon absorption, even the slightest electron-hole band asymmetry causes a transient shift of the chemical potential, causing a disturbance $\delta q$ of the FS. This causes a destruction of the nesting at $k_F$ and a suppression of the divergence of the electronic susceptibility at $2k_F$, causing the electronic charge density wave to rapidly melt\[12\]. A fast destruction time in CDWs is experimentally confirmed: in K$_{0.3}$MoO$_3$, $\tau_m = 50 \sim 100$ fs\[12\] which is an order of magnitude less than $\tau_c \sim$ 0.9 ps in LaSCO, which has a similar $e-p$ interaction strength. Indeed, quite generally, the melting time $\tau_m$ is significantly shorter than the $\tau_c$ in superconductors, as shown in Table I (not all risetimes are reliably measured so far, so only the unambiguous ones are listed). With $\tau_m = 50 \sim 100$ fs, the ions can just barely move into new equilibrium positions within this time, (the 1/4-period of the amplitude mode in K$_{0.3}$MoO$_3$ is $\sim$ 150 fs)\[4\].

We conclude that photon-induced change of state in superconductors and CDW systems relies on very different mechanisms, occurring on different timescales and requiring different amounts of energy. Systematic and experimentally precise measurements of the dependence of $U_c$ on doping and on $T_c$ in cuprates, pnictides, low-$T_c$ superconductors show that vapourisation can be described by the established Rothwarf-Taylor QP recombination mechanism, in a process where energy transfer to the condensate is slowed down and made inefficient by the phonon-QP relaxation bottleneck. In CDW systems on the other hand, the dominant mechanism for the destruction of the ordered state is electronic, so in spite of the fact that the CDW gaps $2\Delta_{CDW}$ are typically much larger than the highest phonon frequencies, and electron-phonon coupling strengths are similar as in cuprates, the destruction is faster and more direct.

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