Fano interference between collective modes in cuprate high-$T_c$ superconductors

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Cuprate high-$T_c$ superconductors are known for their intertwined interactions and the coexistence of competing orders. Uncovering experimental signatures of these interactions is often the first step in understanding their complex relations. A typical spectroscopic signature of the interaction between a discrete mode and a continuum of excitations is the Fano resonance/interference, characterized by the asymmetric light-scattering amplitude of the discrete mode as a function of the electromagnetic driving frequency. In this study, we report a new type of Fano resonance manifested by the nonlinear terahertz response of cuprate high-$T_c$ superconductors, where we resolve both the amplitude and phase signatures of the Fano resonance. Our extensive hole-doping and magnetic field dependent investigation suggests that the Fano resonance may arise from an interplay between the superconducting fluctuations and the charge density wave fluctuations, prompting future studies to look more closely into their dynamical interactions.

Interactions between the intrinsic degrees of freedom of a solid (e.g., charge, spin, orbital, and lattice) often lead to interesting physical properties or phenomena, such as BCS superconductivity, colossal magnetoresistance, and have been key to understanding and predicting novel phases of matter including topological insulators and unconventional superconductivity. Often, the signatures of these interactions manifest in the dynamical response of a system. For example, electron-phonon interaction may significantly renormalize the quasiparticle (electronic) dispersion, introducing kinks into the latter, which has been the subject of extensive research in the case of...
high-$T_c$ superconductors$^{11}$. The same interaction may also introduce discontinuities into the phonon dispersion, a well-known example being the Kohn anomaly arising from an electronic instability driven by Fermi surface nesting. When a discrete excitation interacts with a continuum of excitations, another kind of discontinuity could manifest in its light-scattering amplitude, an asymmetric amplitude profile may develop across the discrete mode, with one side showing a pronounced suppression (a.k.a. anti-resonance) (Fig. 1b). Known as the Fano resonance or the Fano interference, this quantum mechanical phenomenon is closely related to a classical counterpart, i.e., the driven coupled harmonic oscillators model$^4$. In this classical model, if one oscillator is heavily damped (therefore analogous to a continuum of states) while the other is underdamped, the system would exhibit an anti-resonance where the amplitude of the driven oscillation goes through a minimum while its phase undergoes a negative π jump (opposite to the direction of a resonant phase jump) as a function of the driving frequency. In fact, the negative π phase jump is also intrinsic to a Fano resonance, although this particular phase signature has rarely been experimentally evidenced. In this work, we show that the Fano interference is characterized by the asymmetrical line shape of the amplitude response (blue), and also by the negative π jump in the phase response (red), a.k.a. the anti-resonance. Upper panel: in typical spectroscopy experiments, the electromagnetic driving frequency ($\omega_{\text{drive}}$) is swept. Lower panel: in our experiment the driving frequency is fixed, but the resonance frequency of oscillator 1 ($\omega_{\text{osc1}}$) is swept. Here, oscillator 1 represents the superconducting fluctuations, whose energy scale is a function of temperature. By sweeping temperature from 0 towards $T_c$, the energy of the superconducting fluctuations decreases from maximum to zero. We chose the direction of the $\omega_{\text{osc1}}$ axis as shown so that it corresponds to a temperature axis that increases to the right. Note, however, that sweeping temperature is not entirely equivalent to sweeping driving frequency, as additional parameters like the damping constant may depend on temperature. Details of the generalized Fano resonance model can be found in Supplementary Materials S8.

![Fig. 1 | Nonlinear terahertz spectroscopy of d-wave superconductors.](https://example.com/fig1.png)

Fig. 1 illustrates the amplitude oscillation of the superconducting order parameter ($\Delta$) as a function of temperature. A microwave/terahertz-driven superconductor has been known to exhibit strong nonlinearities across $T_c$, particularly in terms of third harmonic generation (THG)$^{13}$. Its origin has been suggested to relate to collective fluctuations inside a superconductor$^{9,13}$, including the superconducting amplitude fluctuations (i.e., Higgs mode$^{14,15}$), the Josephson plasma mode/the nonlinear Josephson current, and also the charge fluctuations (a.k.a. the BCS response, not to be confused with charge density wave fluctuations). In particular, for a conventional superconductor, the energy scale of the Higgs mode and the charge fluctuations follow a similar mean-field temperature dependence as $2\Delta(T)$. They are expected to become resonantly driven when $2\Delta(T) \approx 2\omega$, where $\omega$ is the frequency of the terahertz field. Therefore, it is difficult to distinguish between these different contributions based on the temperature dependence of THG. Nevertheless, for a BCS superconductor in the clean limit, different polarization dependence has been predicted for THG arising from each mechanism (see Supplementary Materials S6 for a detailed discussion)$^{10,11}$, providing a potential way for distinguishing the origin of THG in an experiment.

One caveat about such an experiment is that dissimilar to most spectroscopy experiments in which the electromagnetic driving frequency is swept relative to an excitation fixed in energy, here the driving frequency is kept constant but the energy of the superconducting fluctuations, given by $2\Delta(T)$, is swept by varying temperature (Fig. 2b). For a reasonably small driving frequency $\omega$, as we increase the temperature from 0 the superconducting fluctuations are first driven below resonance (i.e., $2\omega < 2\Delta(T)$) and then above (i.e., $2\omega > 2\Delta(T)$). As illustrated in Fig. 2b, this means that as we heat up a superconductor we expect the THG phase to evolve positively across a resonance and negatively across an anti-resonance. For cuprate high-$T_c$ superconductors where $2\Delta$, according to some experimental interpretations$^{9,10}$, remains sizeable at $T_c$, the resonance of the superconducting fluctuations might move beyond $T_c$ for the small terahertz driving frequency of 0.7 THz used in our experiment. Here, the use of a narrow-band multi-cycle terahertz field generated from a superradiant terahertz source TELBE allows us to precisely extract the phase information required for understanding any resonance or anti-resonance behavior of a periodically driven superconductor.

### Results

Figure 2a shows the typical THG response of a superconducting cuprate thin film. In the raw transmission data, the THG response ($3\omega = 2.1$ THz) is superposed on top of the transmitted linear driving field ($\omega = 0.7$ THz), which can be separated from each other using Fourier lowpass and highpass filters. From their respective waveforms, we obtain their amplitude ($A$) and phase ($\Phi$) by fitting them to the equation

$$E(t) = A \exp(-(t-t_0)^2/c^2) \sin(\omega(t-t_0) - \Phi),$$

(1)

(only $A$, $\Phi$, $c$ are fitting parameters) which allows us to extract the relative phase between the THG response and the linear drive:
In the conventional $s$-wave superconductor NbN ($T_c \approx 14\, \text{K}$, $2\Delta_{0,\text{up}} \approx 5\, \text{meV}$), we observe a peak in THG intensity ($I_{\text{THG}}$) simultaneous with a positive evolution of its phase ($\Phi_{\text{THG}}$) across $T = 10.7\, \text{K}$ (Fig. 2c), which indicates the resonance of the superconducting fluctuations under a periodic drive. The resonance temperature $10.7\, \text{K}$ is roughly expected from the resonance condition $\omega = 2\Delta(T)$, where the driving frequency $\omega$ used for this measurement is $0.5\, \text{THz}$ ($-2\, \text{meV}$) and $2\Delta(T)$ follows a mean field-like temperature dependence. The (optimally doped) cuprate high-$T_c$ superconductors, we observe a slight positive evolution of $\Phi_{\text{THG}}$ at low temperatures (Fig. 3b, see Supplementary Materials SII for results from the bilayer cuprate DyBa$_2$Cu$_3$O$_{7-\delta}$, which has the positive phase evolution at low temperature is more apparent). The phase jump is expected.

Fig. 2a shows the respective waveforms of the driving field and the THG (blue) waveforms are extracted from the raw transmission data (black) using 1.4 THz Fourier lowpass and highpass filters. The peak in $I_{\text{THG}}$ and the continuous evolution of $\Phi_{\text{THG}}$ across $T_c$ indicate the resonance in THG. The error bars on $I_{\text{THG}}$ and $\Phi_{\text{THG}}$ denote the uncertainties in fitting the THG phase and the linear drive phase.

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In addition to the doping dependence, we also looked at the magnetic field dependence of the Fano interference. We measured the THG response of the $x = 0.16$ (OP43) and $x = 0.30$ (OD30) samples under a $c$-axis magnetic field. As shown in Fig. 4, a broadening of the anti-resonance is manifest in both the amplitude and phase response of THG: the dip feature in $I_{\text{THG}}(T)$ disappears while the sharp jump in $\Phi_{\text{THG}}(T)$ widens. Together, these observations suggest that the coupled mode becomes heavily damped or softened upon the application of a magnetic field.

**Discussion**

Below, we offer an interpretation of our results based on a hypothesized interaction between the terahertz-driven superconducting amplitude fluctuations (i.e., Higgs mode) and the CDW fluctuations, which are often found to coexist in the phase diagram of hole-doped cuprates (see Supplementary Materials S7 for a discussion about other possible scenarios for this coupled interaction). Such a hypothesis has also been put forth by previous pump–probe studies focusing on the CDW amplitude fluctuations in underdoped cuprates$^{12}$, and is corroborated by recent reports of ubiquitous charge order fluctuations in a...
large part of the cuprate phase diagram overlapping with the superconducting dome and the pseudogap\cite{28-30}. Note that this interpretation is predicated on assuming THG below $T_c$ predominantly arises from the terahertz-driven Higgs oscillations\cite{31,32} (see Supplementary Materials S5 and S6 for a discussion about the different origins of THG in a superconductor).

To facilitate the discussion, we quickly summarize the current experimental understanding about the CDW/charge order in hole-doped cuprates: (1) the CDW correlation is strongest in underdoped cuprates and weakens with increasing hole-doping or temperature\cite{33,34}; (2) at ambient pressure the CDW correlation is generally two-dimensional and short-ranged at all temperatures and its growth upon cooling is pre-empted by the superconducting transition\cite{35,36}. The second point here implies that any fluctuation is expected to soften because at ambient pressure it sits above the putative critical point inside a generic cuprate superconductor\cite{37,38}. Finally, we note that a recent mean-field investigation of NbSe$_2$ in which superconductivity and CDW coexist and dynamically interact indeed suggests that its THG response manifests a characteristic anti-resonance\cite{39,40}, similar to our experimental observations above (see also Supplementary Materials S10).

While the interpretation of the Higgs-CDW amplitude mode interaction remains speculative at this stage, we note another interesting finding from the magnetic field dependence study: despite significant changes in $I_{3a}(T)$ near $T_n$ (i.e., the suppression of the Fano interference), the overall suppression of THG by the applied field is small up to 10 T in both samples above and below $T_c$. This can be clearly seen for the OD30 sample in Fig. 3c but is more evident for both samples from the field-sweep measurements performed at constant temperatures (see Supplementary Materials S9). As the THG signal is sensitive to small fluctuations and drifts in the driving power (i.e., $I_{3a}$ vs $I_{3a}$) and a complete temperature-sweep measurement as shown in Fig. 3 takes many hours to acquire, the true field dependence of THG is more accurately reflected by the field-sweep results. The lack of field dependence in $I_{3a}$ away from $T_n$ suggests that the length scale relevant to the THG process is more likely related to the superconducting coherence length ($\xi_{ab}$ - several hundred nm) than the London penetration depth ($\lambda_{ab}$ - several hundred nm). The latter is known to increase in the presence of a magnetic field\cite{41,42}, which would have led to a decrease in $I_{3a}$ at any given temperature. The coherence length, on the other hand, is more robust to external fields as it scales with the upper critical field $H_{c2}$ as $\xi_{ab} \sim H_{c2}^{-1/2}$, where $H_{c2}$ is on the order
of several tens of Teslas in these materials. Adopting such an interpretation would immediately suggest that the above-$T_c$ THG, which is also observed in our studies, may arise from local preformed Cooper pairs. In comparison, in NbN THG drops to zero above $T_c$, consistent with the absence of preformed Cooper pairs in conventional superconductors.

The above interpretation carries an important implication as in some of our measurements an above-$T_c$ anti-resonance phase jump may also be inferred (Supplementary Materials S11), implying a non-vanishing interaction between preformed Cooper pairs and CDW above $T_c$. Such an interpretation naturally reminds the intimate relationship between the two orders in the pseudogap region of the cuprate phase diagram. We note that a recent theoretical model, the pair density wave (PDW) model37, predicts a common origin of these two microscopic orders and an interference between Cooper pairs/CDW/PDW already above $T_c$. To investigate the potential link between the PDW model and our findings, and to unlock the longstanding mysteries of high-$T_c$ superconductors, we anticipate future studies to expand our results to greater details and shed further light on the nature of the Fano interference universally manifested by different families of cuprates.

**Methods**

**Sample preparation**
The La$_{2-x}$Sr$_x$CuO$_4$ samples are grown by molecular beam epitaxy (MBE) method on LaSrAlO$_4$ substrate. All samples are 40 nm thick. $T_c$ is determined from mutual inductance measurements as shown in Supplementary Materials S2.

**Experimental method**
The THG experiment is performed using the schematic shown in Supplementary Materials S1. The multicycle terahertz driving pulse is generated from the TELBE super-radiant undulator source at HZDR38. For the data presented in the main text, we use a driving frequency of 0.7 THz and place a 2.1 THz bandpass filter after the sample for filtering the THG response. The transmitted terahertz pulse is measured using electro-optical sampling inside a 2 mm ZnTe crystal and by using 100 fs gating pulses with a central wavelength of 800 nm. The accelerator-based driving pulse and the laser gating pulse have a timing jitter characterized by a standard deviation of ~20 fs. Synchronization is achieved through pulse-resolved detection as detailed in ref. 39.

**Data availability**
Data that support the findings of this study are available from the online repository [https://doi.org/10.14278/rodare.1692](https://doi.org/10.14278/rodare.1692).

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Author contributions
H.C. and S.Ka. conceived the project. H.C., S.Ka., S.Ko., and J.-C.D. developed the experimental plan. Measurements on the cuprate samples were performed at the TELBE facility. S.Ko., H.C., L.F., M.-J.K., H.L.P., K.H., R.D.D., M.C., N.A., A.N.P., I.I., M.Z., S.Z., M.N., J.-C.D., and S.Ka. conducted the beamtime experiment. Measurements on NbN were performed by Z.X.W., T.D., and N.W. at Peking University. H.C. performed data analysis and interpreted the results together with M.B., F.B., M.M., D.M., S.Ka., and B.K. L.S., R.H., M.P., and D.M. performed theoretical modeling. G.K., D.P., G.C., and G.L. provided and characterized the cuprate samples. H.C. wrote the paper with S.Ka., D.M., A.D., L.S., and M.P. with inputs from all authors.

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Competing interests
The authors declare no competing interests.
