An inelastic x-ray study of phonon broadening and charge density wave formation in ortho-II ordered YBa$_2$Cu$_3$O$_{6.54}$

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Inelastic x-ray scattering is used to investigate charge density wave (CDW) formation and the low-energy lattice dynamics of the underdoped high temperature superconductor ortho-II YBa$_2$Cu$_3$O$_{6.54}$. We find that, for a temperature $\sim 1/3$ of the CDW onset temperature ($\approx 155$ K), the CDW order is static within the resolution of the experiment, that is the inverse lifetime is less than 0.3 meV. In the same temperature region, low-energy phonons near the ordering wavevector of the CDW show large increases in their linewidths. This contrasts with the usual behavior in CDW systems where the phonon anomalies are strongest near the CDW onset temperature.

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I. INTRODUCTION

Collective spin or charge density fluctuations are universally present in metals. In some materials they remain dynamic. In others, notably the cuprate high temperature superconductors (HTS), they can become static leading to new ordered states. Charge density waves (CDWs) were recently observed by x-ray diffraction in the high temperature superconductors (HTS) YBa$_2$Cu$_3$O$_y$ (YBCO) and (Y/Nd)Ba$_2$Cu$_3$O$_{4+\delta}$. The existence of ground states with competing order is central to many theories of HTS. A widely discussed example is “stripe order”, that is a state with coexisting charge and spin order.

The CDWs observed in YBCO develop inside the pseudogap phase, and compete strongly with HTS, as evidenced by the reduction in CDW Bragg peak intensity observed on cooling through $T_{SC}$ and the increase in intensity seen on suppressing superconductivity with magnetic field. Signatures of this electronic ordering have been observed by a number of other probes, including scanning probe microscopy, high-field NMR, ultrasound, the Kerr effect, thermal transport, and the Hall effect. The transport coefficients are smoothly connected to the low-temperature high-field quantum oscillations (QOs) that demonstrated the existence of small Fermi surface (FS) pockets. However, some of these measurements only show signs of ordering at high fields, in contrast with the x-ray scattering experiments, which observe the CDW in zero field.

The x-ray diffraction measurements reported to date have been performed with very few or no broad energy analysis. Thus the reported wavevector dependent scans integrate over both Bragg peaks and excitations such as phonons. As is discussed elsewhere, NMR and ultrasound probe much lower frequencies than conventional x-ray diffraction. It is possible that the correlations involved in the CDW become “quasi-static” (slow) and visible to x-rays before they are seen in NMR and ultrasound. Thus it is important to make diffraction measurements with higher energy resolution.

In this article, we report a study of the CDW ordering and the low-energy phonons in ortho-II (YBa$_2$Cu$_3$O$_{6.54}$) using inelastic x-ray scattering (IXS) with an energy resolution of 1.5 meV. We find that (i) the CDW is ordered on an energy scale less than the energy resolution of the experiment; and (ii) there is a strong broadening of the lowest energy phonon linewidth at the ordering wavevector of the CDW, which sets in on entering the CDW state.

II. BACKGROUND

Charge density waves are not uncommon in metals (see Ref. [15] for a recent review). Some of the most well-known examples are the transition metal chalcogenides, such as 2H-NbSe$_2$ and TaSe$_2$. A charge density wave is essentially the formation of a periodic modulation of the electron density, and is typically associated with a periodic lattice distortion (these may or may not be commensurate with the crystal lattice). This electron (or charge) density modulation may be brought about by electron-phonon or electron-electron interactions.

Structure in the charge response function of a metal is reflected in the phonon energies, via the electron-phonon interaction, this will give rise to Kohn anomalies in the phonon spectrum associated with the Fermi surface. The response function of a 1-D electron gas diverges at a wavevector $2k_F$ (twice the Fermi wavevector). For this case, the Kohn anomaly will occur at a wavevector of $2k_F$, and if the system exhibits a Peierls instability, the Kohn anomaly will grow, with the phonon frequency eventually softening to zero at a given temperature (Fig. 1a). At this point the CDW has stabilized - the phonon has “frozen in”. This has been observed in 1-D systems such as K$_2$Pt(CN)$_4$Br$_{0.3}$,3.2 D$_2$O (KCP) and K$_{0.3}$MoO$_4$.

In higher dimensions, if the same simple physics applies to the electron gas, the renormalization of the phonon frequen-
cies at the Fermi surface is less severe, and a phase transition to the frozen-in state is not expected. However, strong Kohn
anomalies (with \( \omega_{\text{ph}} \rightarrow 0 \)) have also been observed in higher
dimensional systems, such as the 2-D system 2H-NbSe\(_2\). In
cases such as these, an additional wavevector dependence
must exist, either in the electronic response function \( \chi_q \) or in
the electron-phonon interaction, and this is sufficient to freeze
in the relevant phonon, giving rise to the (static) CDW.

There are also several examples of CDWs developing in
systems where \( \omega_{\text{ph}} \rightarrow 0 \), e.g. NbSe\(_2\) and (TaSe\(_4\))\(_2\). In
these materials, there may be some phonon softening, i.e. a
shallow anomaly in the dispersion, and an increase in the
phonon linewidth is observed. In this case, the simple picture
of the phonon freezing to give the CDW is not valid. We note
that in all the systems described above, phonon anomalies are
strongest near the onset temperature of the CDW, \( T_{\text{CDW}} \).
In view of the variety of CDW behavior it is important to establish
which class of system YBCO belongs to.

III. EXPERIMENTAL DETAILS

We carried out (23.724 keV) x-ray scattering experiments
at the XOR 30-ID HERIX beam line\(_{32,33}\) at the Advanced
Photon Source, Argonne National Laboratory. HERIX allows
the x-ray cross-section to be measured as a function of momentum \( Q = k_i - k_f \) and energy transfer \( E_f - E_i \) of the
photon. The scattered beam was analysed by a set of spheri-
cally curved silicon (12 12 12) analysers. The full width
at half maximum (FWHM) energy resolution was 1.5 meV.
The sample was mounted in a closed cycle cryostat on a 4-
circle goniometer. The experiment was performed in trans-
mission geometry. Inelastic measurements were made in con-
stant wavevector mode. We measured the elastic line during
each scan to correct for any drifts in energy calibration.

The sample used was a \( \sim99\% \) detwinned YBa\(_2\)Cu\(_3\)O\(_{6.54}\)
single crystal of dimensions \( 1.5 \times 3 \times 0.16 \) mm (the
same sample used in Ref. 4). YBa\(_2\)Cu\(_3\)O\(_{6.54}\) differs from,
e.g. La\(_{2-x}\)(Ba,Sr)\(_2\)CuO\(_4\), in that it contains bilayers of CuO\(_2\)
planes, separated by layers containing a certain fraction (de-
pending on \( x \)) of Cu-O chains. The oxygen-filled chains,
which run along the orthorhombic crystal b-direction tend
to order and are labelled ortho-N, depending on the repeat
length (\( Na \)) of the ordering of the chains along \( a \). This
sample was of the type ortho-II, with alternate Cu-O chains
occupied. The lattice parameters are \( a \approx 3.82, b \approx 3.87, \)
and \( c \approx 11.7 \) Å (ignoring the chain-ordering superlattice),
\( T_{\text{CDW}} = 155 \pm 10 \) K and \( T_{\text{SC}} = 58 \) K (as measured on a
Quantum Design MPMS magnetometer). Following previous
practice\(_4\), we label reciprocal space using reciprocal lattice
units \( (2\pi/a, 2\pi/b, 2\pi/c) \) ignoring the additional periodicity
introduced by the chain ordering.

The CDW produces incommensurate satellite Bragg peaks
at positions \( Q = \tau \pm q_{\text{CDW}} \), where \( \tau \) is a reciprocal lattice point
of the original (undistorted) lattice and \( q_{\text{CDW}} \) is the wavevec-
tor of the CDW. The CDW in ortho-II YBCO has two funda-
mental wavevectors, \( q_1 = (\delta_1, 0, 0.5) \) and \( q_2 = (0, \delta_2, 0.5) \),
where \( \delta_1 = 0.320(2) \) and \( \delta_2 = 0.328(2) \) (Ref. 4). Previous
hard x-ray measurements have indicated that the intensity of
the CDW satellite Bragg peaks is strongly dependent on \( \tau \),
with \( Q = (0.2 - \delta_2, 6.5) \approx (0, 1.672, 6.5) \) being a particularly
strong peak. We therefore concentrated on measuring near
this position.
IV. RESULTS

A. Charge Ordering

Fig. 2 shows contour plots based on a series of constant wavevector scans made in the region of $Q = (0, 2 - \delta_2, 6.5)$. The data were collected at $T = 55 \pm 2 \text{ K} \approx T_{SC}$ and $T = 148 \pm 2 \text{ K} \approx T_{CDW}$. A temperature near $T_{SC}$ was chosen because the previous measurements show that the satellite intensity is maximal at $T_{SC} \approx 148 \text{ K}$. The $T = 148 \text{ K}$ data (right panel) shows a ridge of elastic scattering centered on $E = 0$ meV. This background increases towards the $(0, 2, 6.5)$ position and is due to disorder in the sample, frozen in above room temperature. On cooling to $T = 55 \text{ K}$, a peak develops near $Q \approx (0, 1.675, 6.5)$ corresponding with the CDW ordering. This peak is on a sloping background (no background corrections have been made to the data).

Fig. 3 shows individual scans through the CDW as a function of wavevector and energy. The value of $q_2$ measured is $0.327(2)$ r.l.u., in agreement with the values reported in Ref. 4. The peak is resolution-limited in the wavevector scans. Comparing the energy dependent scans at 155 K and 55 K, we are able to fit the additional scattering due to the CDW Bragg peak using a Gaussian function with a FWHM of $\Delta E_{\text{FWHM}} = 1.46(10)$ meV. This is indistinguishable from our estimate of the experimental energy resolution (1.5 meV), within experimental error.

We determined the temperature dependence of the CDW Bragg peak from a series of energy dependent scans (as in Fig. 3(b)) through the $(0, \delta_2, 6.5)$ position. In order to obtain a good fit of the lineshape we used two Gaussian functions centered on $E = 0$ (plus a constant background), to provide a better fit of the tails of the peak. For example, the data in Fig. 3(b) gave widths $\Delta_1 \approx 1.46$ meV and $\Delta_2 \approx 2.02$ meV. Fig. 4 shows the temperature dependence of the sum of the two peak heights (i.e. the $E = 0$ response). The Bragg peak intensity follows our hard x-ray data. This was collected without energy analysis, and therefore integrates over a range of energies up to $\sim 1$ keV, orders of magnitude greater than the $\approx 1.5$ meV resolution of the present measurements. The dotted line in Fig. 4 shows the $T$-dependence from our previous experiment, which appears to be consistent with the present experiment. As the temperature is lowered, the Bragg peak intensity starts to increase at $T = 155 \text{ K} \approx T_{CDW}$. The highest intensity is measured at $T = 55 \text{ K} \approx T_{SC}$. As the temperature is lowered further the competition between superconductivity and the CDW causes the intensity to reduce.

B. Phonon anomalies

We made energy dependent phonon scans for $T = 55$ and 155 K at various positions along $(0, k, 6.5)$ for $1.5 \leq k \leq 0$ near the strong $(0, 2 - \delta_2, 6.5)$ CDW satellite Bragg peak found in our previous study. Fig. 5(a)-(h) shows energy scans for wavevectors near the ordering position. Data such as that in Fig. 5(a)-(h) may be used to determine the phonon dispersion curves; these are shown in Fig. 5(i)-(j). Fig. 5 shows that the lowest energy phonons near the ordering wavevector are strongly renomalized on entering the CDW state. Specifically, if we compare the scans at the ordering wavevector $q_2$ - panels (b) and (f) - we see that the line shape of the phonons broadens on cooling from 155 K to 55 K. This means that spectral weight appears at lower energies (see Fig. 5), and there is a drop in the peak intensity of the phonons. Similar effects are also observed in panel (c). If we follow the peak intensity across the series (a)-(d), scans (b) and (c) show these anomalies most clearly. The dispersion curves in Fig. 5(i)-(j) were obtained by fitting multiple Gaussian lineshapes to the spectra. The points show the peak positions and vertical bars show the resolution-deconvoluted (FWHM) widths of the phonon peaks. In most cases, the widths are less than the point size. However, the big increase in width near $q_2$ at low-
temperature is obvious.

Fig. 6 shows fits of the data in Fig. 5(b, f) to a damped harmonic oscillator response (for each phonon mode),

\[
\chi''(\omega) = \frac{\gamma}{(\omega^2 - \omega_0^2)^2 + (\gamma/2)^2}.
\]

where \(\gamma = \gamma_0\) and the phonon frequencies, \(\hbar\omega_i\), are \(\hbar\omega_{1}\) = 8.7 ± 0.1 meV and \(\hbar\omega_{2}\) = 10.6 ± 0.3 meV. Setting the damping factor \(\gamma\) for the two modes to be equal, the response at \(T = 155\) K can be explained (see Fig. 6) with \(\gamma = 4.2 \pm 0.8\) meV and unchanged values for \(\hbar\omega_{1}\) and \(\hbar\omega_{2}\). Within this phenomenology, the damping introduces a small shift in the peak of the response (Eqn. 1), \(\Delta\omega \approx -\gamma/8\omega \approx 0.3\) meV which is not directly discernible within the resolution of with experiment.

V. DISCUSSION

A. Charge Ordering

Charge density waves have rather unique dynamical properties. It is well known that CDWs can be easily unpinned from the crystal lattice by the application of an electric field leading to so-called sliding charge density waves (SCDW)\(^{25}\). In the case of ortho-II YBCO, the “charge ordering” anomalies seen in NMR\(^8\) and ultrasound\(^10\) occur at lower temperatures and high magnetic fields (Table I) than observed with

100 keV x-ray diffraction.

This has led to speculation\(^{10}\) that the conventional x-ray diffraction experiments\(^{1–5}\) are, in fact, observing “dynamic correlations”. Our IXS measurements are carried out with energy discrimination and therefore allow us to put a bound on the temperature of the ordered state. We estimate\(^9\) that, at \(T = 55\) K, the (deconvolved) FWHM width (\(\Delta\)) of the CDW Bragg peak is \(\Delta < 0.3\) meV. This puts a lower bound on the lifetime of \(\tau \approx 2\hbar/\Delta \approx 4.4\) ps. Thus the frequency scale (\(\hbar\omega_{CDW} \approx 0.3\) meV) corresponding to the CDW order is at least two orders of magnitude less than that of the superconducting gap \(2\Delta_{SC} \approx 30\) meV and pseudogap (\(\Delta_{PG} \approx 100\) meV)\(^{29}\).

Returning to the comparison with NMR and ultrasound measurement on ortho-II YBCO, these are both low frequency probes, with an energy scale of 1.5 \(\mu\)eV for the NMR. It is possible that there is an additional phase transition at lower temperatures to a “frozen” CDW state seen by these probes. An alternative possibility is a lock-in transition to commensurate ordering, that is, \(\Delta_2 \rightarrow 1/3\). Such lock-in transitions are common in CDW systems\(^{16–19}\), and are accompanied by anomalies in the elastic constants\(^{21,22}\) and therefore we deem this more likely.

B. Phonon anomalies

Charge density waves are closely connected with phonon anomalies. As discussed in Sec. II, 1-D CDW systems\(^{15,19}\) show phonon softening or “Kohn anomalies”, where the frequency of the phonon associated with the CDW is reduced to zero. In less 1-D systems, the anomalies are usually weaker, however, we note the dramatic softening of the phonons recently observed in the well-known CDW material 2H-NbSe\(^{25}\). The present results are similar to materials such as NbSe\(^{21}\) which show a large increase in the phonon linewidth near the CDW ordering wavevector. The difference between present data on YBa\(_2\)Cu\(_3\)O\(_{6.5}\) and other CDW systems\(^{18,19,21,22,23}\) seems to be the temperature at which the broadening in the phonon linewidths occurs. We observe close to resolution limited linewidths near \(T = 155\) K \(\approx T_{CDW}\) for the phonons

| Probe                  | \(E_{probe}\) (\(\mu\)eV) | \(T_{CO}(K)\) | \(B(T)\) |
|------------------------|--------------------------|----------------|---------|
| NMR\(^8\)             | 1.5                      | 50             | 28.5    |
| NMR\(^9\)             | 0.5                      | 60             | 10.4    |
| ultrasound\(^10\)     | 0.6                      | 44.8           | 26.4    |
| 0.931 keV x-ray diffraction\(^10\) | 130 meV         | 150            | 0       |
| 100 keV x-ray diffraction\(^10\) | 1 keV               | 155(10)        | 0       |
| IXS (this work)       | 1.5                      | 150(40)        | 0       |

### Table I

Charge ordering (CO) temperatures in ortho-II YBCO observed by various probes and the energy scale on which the charge correlations are probed (\(E_{probe}\)). In the case of NMR and ultrasound \(E_{probe}\) is taken as the energy at which the charge response is probed. For the diffraction measurements, it is the energy range of integration i.e. the energy resolution of the instrument.
Resolution. (i)-(j) Phonon dispersion curves along the wave structure of the pseudogap properties in the normal state by the pseudogap. Thus, the such as the modification of the low-energy electronic properties controlled by something other than the superconducting gap (charge) response of the conduction electrons. The onset of phonon linewidths (and energies) are affected by superconductivity. On cooling conventional superconductors through $T_{\text{SC}}$, the damping is reduced for low-energy phonons, while for higher energies $\sim 2\Delta$ it can increase. This change arises because of the modification of the screening (charge) response of the conduction electrons. The onset of extra damping above $T_{\text{SC}}$ in YBa$_2$Cu$_3$O$_{6.4}$ indicates that it is controlled by something other than the superconducting gap such as the modification of the low-energy electronic properties in the normal state by the pseudogap. Thus, the $d$-wave structure of the pseudogap may mean that the damping is increased for the energy ($\sim 8$ meV) and wavevector $[\mathbf{q}=(0,0.328,0.5)]$ of the wavevector of the anomalous phonon studied here.

We note that a new collective low-energy ($Q=0$) mode, which appears approximately at $T_{\text{CDW}}$, has been observed by time-resolved reflectometry in underdoped ortho-III and ortho-VIII YBa$_2$Cu$_3$O$_{6+x}$. It shows marked changes at $T_{\text{SC}}$. The mode has frequency $\sim 7.7$ meV, close to that of the damped phonons reported here. It would also have components at $\mathbf{q}=q_{\text{CDW}}$ and therefore may interact with a phonon at this wavevector and energy.

The phonon cross-section of IXS depends on $(Q \cdot \epsilon)^2$, where $\epsilon$ is the polarisation of the phonon mode. At the $Q=(0.2-\delta,6.5)$ position, the scattering vector is $52^\circ$ from the $b^*$ axis, and so our measurement is approximately equally sensitive to atomic motion in the $b$ and $c$ directions. Our earlier diffraction work has shown that the CDW modulation has modulations along both these direction, and so we expect that the broadened phonon has displacements along the same direction. A recent DFT calculation suggests the stronger and lower energy mode studied here is the $B_1$, transverse acoustic phonon.

Phonon anomalies have been observed near the $h=0, k=0.3$ position at higher energies (40–55 meV) in YBCO and La$_2-x$Sr$_x$CuO$_4$. These anomalies are associated with oxygen stretching and bending modes, rather than the acoustic mode of the present study. What is interesting is that many phonon branches show an anomaly with the same in-plane component of the wavevector. This suggests that the anomalies stem from the underlying $q$-dependent electronic

![FIG. 5. (a)-(h) IXS $E$-scans of the low energy phonons for wavevectors along the $(0,k,6.5)$ line. Solid lines are fits to a sum of Gaussian functions. Data have been multiplied by $1-\exp(-E/(k_BT))$ to correct for the Bose factor. The horizontal bar in panel (a) is the instrumental resolution. (i)-(j) Phonon dispersion curves along the $(0,k,6.5)$ line for $T=55$ and 155 K. The solid circles represent the phonon peak positions determined from fitting data such as that in (a)-(h); the dashed lines are guides to the eye for the different branches. The resolution-deconvolved phonon widths are represented by vertical bars. The vertical dotted line is the CDW ordering wavevector.](image-url)

![FIG. 6. IXS $E$-scan of the low-energy phonons at $Q=(0.1,672,6.5)$ for $T=55$ and 155 K. Phonons are fitted to a damped harmonic oscillator (DHO) response function (solid lines). Fits are convolved with the instrumental resolution.](image-url)
dielectric function $\chi(q)$ of the CuO$_2$ planes. Evidence to support this scenario comes from STM measurements on Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ which show modulations in conductance maps with similar wavevectors to the one observed here. Some studies above$^{39,40}$ find anomalies to be present in the $k$ direction, but not the $h$ direction — we do not have the data to comment on this at present.

Chan and Heine$^{41}$ have proposed a criterion for a stable CDW phase. Within their picture, CDWs are stabilised either by singularities in the $q$-dependent electronic dielectric function $\chi(q)$ or by a strongly $q$-dependent electron-phonon coupling (EPC). Given the evidence for singularities in $\chi(q)$ presented above, the first scenario of Chan and Heine seems more likely. This conclusion is strengthened by our observation$^4$ that the $q$-vectors of the CDWs vary systematically with doping in a manner that indicates a connection with the electronic states near the Fermi surface.

VI. CONCLUSION

We have carried out an IXS study of CDW Bragg peak and the low-energy phonon modes in the underdoped cuprate ortho-II YBa$_2$Cu$_3$O$_{6.54}$. The CDW order is static within the experimental resolution and we can place an upper bound of 0.3 meV on the energy width of the Bragg peak. This is about two orders of magnitude lower than superconducting gap and the pseudogap. The lowest energy phonon modes show a strong increase in linewidth near the ordering wavevector of the CDW. This results in the appearance of spectral weight at lower energy transfers on lowering the temperature through the CDW onset temperature. Unusually, the phonon broadening is largest at $T_{SC}$ (i.e. $\ll T_{CDW}$) in contrast with the usual CDW behavior.

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In the SI of Ref. 2, we argued that the correlation length of the CDW $\xi \approx 90 \, \text{Å}$ and speed of sound $v_s = 4.6 \times 10^3 \, \text{ms}^{-1}$ can also be used to estimate an upper bound energy (frequency) scale as $\hbar v_s \xi^{-1} \approx 0.3 \, \text{meV}$. 

Ref. 3 studied YBa$_2$Cu$_3$O$_6$. 

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