POSSIBLE LINKS BETWEEN ANOMALIES IN THE PHOTOEMISSION, OPTICAL AND PHONON SPECTRA OF HIGH-Tc CUPRATES

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Abstract

The impressive number of anomalies discovered in high-Tc cuprates by different experimental probes continued to increase also in the last years, adding more to the complexity of the general picture. An intriguing feature of this picture is that the energy scales of these phenomena are very different, spreading from meV to eV. It might imply that the phenomena are actually unrelated. However, the fundamental concept of strong electron correlations, in which high and low energy scales are closely connected, could account in a natural way for the underlying physics and provide an appropriate framework for its description. In this context, the pioneering works of academician V.A. Moskalenko in the last decades constitute a major contribution to the development of this area of physics and our understanding of the emerging new materials.

Being guided by this concept we present arguments for the existence of internal connections between apparently unrelated anomalies reported in photoemission, optical and neutron scattering experiments on high-Tc cuprates: low energy kink, high energy anomaly, softening of the bond-stretching phonon mode and mid-infrared incoherent absorption peak. Our approach is based on both microscopic and semi-phenomenological models and suggests new kinds of experiments that could be of importance for an even broader class of materials.

High values of the superconducting transition temperature (initially around 40 K in the La2−xFxSrxCuO4 and presently over 160 K in Hg0.2Tl0.8Ca2Ba2Cu3O8 compared to 20 K of the highest temperature “classical” superconductor Nb3Sn), magnetic fields (dozens of Tesla) and huge electric currents ~ 10^11 A/m^2 supported by high-Tc cuprates are at the origin of multiple and constantly growing range of applications. Magnetic resonance imaging in medicine, control magnets in particle accelerators and fusion reactors, electromagnetic filters, power cables, power storage, motors, magnetic levitation devices, ultrasensistive detectors – to mention a few. In some cases production costs have become comparable to traditional materials, e.g., coated YBa2Cu3O7 conductors versus copper cables. On the other hand, the potential of high-Tc materials for future applications is promising as well. Since superconductivity is a phenomenon of quantum coherence, these materials are natural candidats for circuit units of future quantum computers, spintronic and nanoelectronic devices. While there are hundreds of high-Tc compounds, they all share a layered structure made up of one or more copper-oxygen planes. The other important fundamental differences from “low temperature” superconductors consist in a different symmetry of the superconducting condensate wave function (d_{x^2−y^2}) and in the Mott-Hubbard (strongly correlated) nature of the parent insulating state [1]. According
to band theory, the band is half-filled and must be metallic. Nevertheless, there is a strong repulsive energy cost when putting two electrons/holes on the same ion. When this energy dominates over the kinetic energy, the ground state is an (Mott) insulator due to strong correlation effects. Superconducting samples are obtained by light chemical doping of the parent compound with holes or electrons. This leads to a very low density of charge carriers and a reduced screening of ionic potential, so the normal state of the material is a very poor metal (sometimes called „strange metal”). The optimal conditions are achieved at small doping ($x=0.15$) and superconductivity disappears beyond 0.25 (overdoping). Charge inhomogeneity is also an important feature that has been reliably established in experiments, e.g., by scanning tunneling microscopy (STM).

Understanding of the underlying principles of high-temperature superconductivity turned to be a challenge whose complexity far exceeds that encountered with previous application-relevant electronic materials. One can say, following P.W. Anderson, that after more than 20 years since the high-Tc discovery the only consensus that has been reached so far is that there is no consensus. The spectrum of microscopic theories ranges from extensions of the „classical” BCS theory [2, 3] to the denial of its basic principles [4], from the fundamental paradigm of Landau Fermi liquid [5] to the marginal Fermi liquid [6] or Luttinger liquid, inspired by known results on one-dimensional models [7], where the fundamental notion of quasiparticles is dismissed by spin and charge separation.

Superconductivity in conventional superconductors such as Pb or Nb is explained by formation of pairs of the charge carriers due to coupling to lattice vibrations. In the cuprate high-Tc superconductors the formation of such singlet pairs has been demonstrated but the bosonic “glue” for their formation is not known. In addition, the very idea of a “glue” is under criticism [8]. The number of existing microscopic theories and concepts is bewildering and it is impossible to give even a brief account of the present status. Nevertheless, phonons remain one of the major candidates for being an essential component of the physics, with different branches of the lattice oscillations being discussed in this context [9]. Another major candidate is a collective spin excitation, the magnetic resonance mode, [10]; there exist theories that consider the charge degrees of freedom and particular dielectric properties to be responsible for the superconductivity [11]. It cannot be excluded that there exists also an intrinsic connection between some of these factors that is important.

It is well known that the metallic state above the superconductor is highly unusual and superconductivity is only one aspect of a rich phase diagram, which must be understood in its totality. Existence of pseudogap [12] in single-particle spectrum till rather high temperatures $T$, the linear in $T$ (instead of quadratic) resistivity [13], temperature dependent Hall coefficient [14], non-Korringa nuclear-spin relaxation rate, anomalous doping-dependent isotope shift [15], violation of sum rules and large amount of spectral weight present in the “intragap” region seen in optical conductivity [16], large featureless temperature independent continuum extending to about 0.5 eV in the Raman scattering [17], large softening of the half-breathing in-plane phonon mode, unaccounted by ab-initio LDA calculations [18], etc. At the same time, cuprates have arguably become the best studied materials outside the semiconductor family. Over the last 10 years an unprecedented progress has been achieved in the development of experimental techniques, including angle-resolved photoemission spectroscopy (ARPES), spectroscopic imaging-scanning tunneling microscopy (SI-STM), microwave/terahertz/infrared/optical spectroscopies, resonant and inelastic x-ray spectroscopy, high intensity neutron scattering (NS), and nuclear magnetic resonance/nuclear quadrupole resonance/muon spin relaxation (NMR/NQR/μSR).
Recent ARPES experiments have revealed a number of new anomalies in the single particle electronic spectra of high-Tc cuprates (high energy anomaly, low energy kink, large isotope effect for states well below the Fermi energy). It raises a natural question about possible connection to the phenomena known before (e.g., large doping and momentum dependent spectral weight transfer, anomalous phonon softening, broad optical absorption peak and electronic Raman continuum within the charge transfer gap and others). We will try to follow the idea of a possible mutual dependence of such apparently not connected phenomena and to propose a model framework to its theoretical description. Quantitative agreement with latest experiments (ARPES, EELS, optical conductivity, neutron and x-ray scattering and others) will be an essential requirement. The novelty of our approach is that it incorporates phenomena of different nature (single particle and collective excitations, spin and charge, electron density and lattice oscillations, coherent and incoherent) and energy scale (from Fermi surface to 1.5 ev) into a unified setting. Motivation for such a possibility follows from our earlier studies where links between some anomalies have been singled out [19-22]. Moreover, we will explore the possibility that similar connections can exist in a broader class of compounds (metallic perovskites) relevant for applications, e.g., colossal magnetoresistance manganates, ruthenates with coexisting ferromagnetism and superconductivity, superconducting bismuthates, nickelates, etc. On the other hand, these materials are believed to be in a “regime” of strong electron correlations, when a single-particle description of elementary spectrum is insufficient.

The so called “low energy kink” in the electron dispersion found in ARPES [23] (see Fig. 1), and the related “peak-dip-hump” structure of the spectral density have been first discovered in high-Tc cuprates and attracted much attention. These phenomena have been mainly associated to the effect of a bosonic mode with energy about 40-70 meV interacting with electrons. The two candidates known to exist in this energy interval have been explored in this context: phonons and the magnetic resonance mode. Later kink structures have been discovered in other materials of the perovskite type (manganates) [24].

In [21, 22] we have considered a microscopic model (Hubbard-Holstein) where the phonon characteristics were taken from experimental data. Our treatment has been based on the approach of V.A. Moskalenko [26-38] developed in [39]. This has allowed to explain a number of anomalous properties, such as the isotope effect of the electronic spectra also at energies much larger than the afore mentioned interval; the shape of the spectral density, its dependence on doping, temperature, and momentum in the Brillouin zone; the unusual doping dependence of the kink structure, when the lower energy region is almost independent on doping, in contrast to the higher energy one and others. In addition to the good quantitative agreement with the experiment, we have also predicted a change in topology of the surface in the Brillouin zone defined by the position of the kink from hole-like to electron-like. The value of the electron-phonon coupling constant following from our analysis turned to be rather large (λ ~ 1) and, therefore, we have also considered the renormalization due to vertex corrections. However, another important implication is that the phonon spectrum should be strongly affected by the interaction with electrons as well.

This brings us to the problem of another anomaly, that of the phonon spectra, which was indeed reported earlier for the high-Tc cuprates. Remarkably, similar anomaly was reported for a number of other materials representative of the perovskite family. Figure 2 shows the oxygen ions oscillation pattern belonging to this mode and Fig. 3 shows examples of its anomalously large softening observed in different materials.
Fig. 1. Electron dynamics in the La$_{2-x}$Sr$_x$CuO$_4$ system, from Ref. [25]. a) Energy $E$ as function of momentum $K$ for various doping $0.03 < x < 0.3$ measured at temperature $T=20$K along the (0,0)-(Pi,Pi) nodal direction. Dispersion is obtained by fitting momentum distribution curve MDC of the ARPES. Arrows indicate position of the kink (about 70 meV) which separates regions with different slope b) scattering rates, full width at half maximum of MDC at $x=0.063$ and $T=20$ K.

Fig. 2. The in-plane CuO$_2$ bond-stretching phonon mode has a natural strong coupling to the in-plane charge carriers relevant for the physics of cuprates. Its dispersion softens towards the edge of the Brillouin zone that can reach 40%; a similar pattern is observed for the increase of the linewidth (see Fig. 3, from [40]).

The previously proposed explanation [41] is based on microscopic t-J model where the calculated charge response function has a peak almost in resonance with the phonon causing the softening. However, the existence of such charge response singularity at the specified energies has not been experimentally established so far, it also raises the question why other phonon modes in the same energy scale are not affected by this resonance and only the highest energy BS mode is affected so hard. We mention that a number of other microscopic models of this phenomenon exist in the literature.

The idea advanced in our paper [20] was qualitatively different: to “read” electronic properties from those of phonons, since the latter can be measured experimentally. Next, we have formulated a rather general ansatz for the charge response function that defines the self-energy of the phonons in terms of damped oscillators. This ansatz complies with the general principles for the response functions (Kramers-Kronig relations, the correct symmetry $\omega \rightarrow -\omega$, e.g., a Lorentzian does not). As is also well known, oscillators form the basis of the Leh-
man representation for a generic bosonic Green function, in this case of electronic origin, i.e., describing density fluctuations. Together with the Dyson equation for the phononic Green’s function, the ansatz can serve as a workable semi-phenomenological model. Thus, for a single oscillator ansatz

\[ D(q, \omega) = \frac{\omega_q}{\omega^2 - \omega_q^2 \left(1 + \alpha_q \sin(qx/2)\right) P(q, \omega)} \]

\[ P(q, \omega) = \frac{\eta_q}{\omega^2 - \Omega_q^2 + i\Gamma_q\omega} \]

where “sin” is the structure factor of the BS mode along the x-axis taken as an example; \( \Omega \) (\( \Gamma \)) is the frequency (damping) of the electronic oscillator, and \( \omega_q \) is the phonon frequency.

Fig. 3. Comparison of the phonon anomaly in the bond-stretching branches observed in different metallic oxide perovskite materials along different directions within the Brillouin zone. Non-stoichiometric La\(_2\)NiO\(_4\), La\(_{1.69}\)Sr\(_{0.31}\)NiO\(_4\), La\(_{1.85}\)Sr\(_{0.15}\)CuO\(_4\), YBa\(_2\)Cu\(_3\)O\(_6\), HgBa\(_2\)CuO\(_4\), Ba\(_{1-x}\)K\(_x\)BiO\(_3\) and La\(_{1-x}\)Sr\(_x\)MnO\(_3\) [40].

However, it appears to be too many parameters in this model, even if one absorbs the weight of the oscillator \( \eta \) into the coupling constant \( \alpha \) further denoted by \( \beta \). Moreover, we would like to have a model, which does not reduce to just fitting the experimental data and instead allows verification of its consistency. To achieve this we propose that the phonon spectrum be measured for isotopically substituted compounds. As the mode is formed by oxygen oscillations, one should consider the rather common O\(^{16}\) - O\(^{18}\) substitution. Then, it turns out that the main requirements mentioned above are fulfilled and the model can be used for quantitative evaluation (see [20] for more details). Such experiments have not been performed before, as one
usually is interested in the isotope effect on electronic properties. At the present stage, we can use the simplified version of the model (i.e., neglecting the q-dependence of the parameters) to see whether the characteristics of the electronic mode derived from the phonon spectrum can be put into correspondence with independent measurements by other probes, e.g., from optical absorption. One then obtains a simple formula

\[ \gamma_q = \Gamma_q \frac{\omega_0^2 - \omega_q^2}{\Omega_q^2 - \omega_q^2}, \]

where \( \gamma \) stands for the phonon damping. Note that this formula does not reduce to simple fitting because we are now confronting two independent sources of data to check whether equality is satisfied and this is not guaranteed from the outset. Then, it follows that the only candidate that comes out is the famous mid-infrared (MIR) anomaly with \( \Omega \leq \Gamma = 0.3-0.6 \text{ eV} \), lower values corresponding to samples with larger doping. This typical structure of an overdamped oscillator often appears in experimental works describing optical conductivity spectra in cuprates. Thus, we can conclude that, instead of the resonance with a narrow lightweight electronic density fluctuation mode, the phonon anomaly is due to an out of resonance broad and much “heavier” MIR excitation. Its spectral weight is known to be comparable to Drude (low energy) part of optical conductivity.

It should be noted that although the quantitative match of the above estimates to the right scale of energies is remarkable, a more detailed analysis outlined earlier is necessary (given the suggested experiments are carried out to allow the consistency tests). Our estimates were based on several simplifications, e.g., the electronic mode being non-dispersive. In particular, at short wavelengths a lower energy electronic oscillator should also be included into the model to account for the detailed structure of the phonon softening closer to the middle of the Brillouin zone (see [42]).

Fig. 4. Intensity of ARPES spectra energies below \( E_{\text{Fermi}} \) in Pb-Bi 2212, from Ref [43]. The white surface to the top-right represents the dispersion of the ARPES peak in the Brillouin zone \( K_x-K_y \). Below is the vertical cross-section showing the spectral intensity versus energy and in-plane momentum. The energy scales are \( E_0 \) and \( E_1 \), the low and high energy kinks, \( E_2 \), the bottom of the waterfall region.
The origin of the highly incoherent (and therefore short lived) MIR electronic excitation is a matter of long lasting debate. Although it is well documented in various optical experiments, nothing is known about its dispersion, since such relatively low energy scale is inaccessible by existing momentum resolved probes. Therefore, the approach described here provides a unique opportunity to study dispersive electronic density excitations at low energies. In this case phonons can be used as a probe for the excitations they are coupled to.

The relevance of the intrinsic connections between the excitations described above has been recently reinforced by the discovery of the so called “High energy anomaly” (HEA) in ARPES spectra [43, 44] (see Fig. 4).

The anomaly is composed of a new kink that appears around 0.35-0.4 eV and is followed by a sharp drop ("waterfall") of the spectral intensity from 0.4 till 0.9-1 eV at almost no change in momentum, to regain the parabolic dispersion of the "bare" electronic band. The spectral weight involved in HEA is both orders of magnitude larger than that of the low energy kink, and highly incoherent, i.e., the linewidth of the ARPES signal in the waterfall area is comparable to the energy itself. The flattened quasiparticle part of the spectrum (above $E_0$) is due to correlation effects (band narrowing or heavier effective mass) and, therefore, the existence of "waterfalls" should have different origins, although [45] claims otherwise. Until recently the HEA was also not detected in the numerical simulations on fundamental models of high-Tc such as Hubbard, Emery, or t-J models [46]. Several other scenarios have been already proposed, among those the most “popular” is related to magnetic degrees of freedom, namely the spin-wave spectrum that is governed by the exchange interaction $J$ about 0.15 eV. This spectrum, however, can be directly extracted from polarized neutron scattering, including the distribution of spectral weight, etc. Then calculations show that the magnetic "boson" does not reproduce quantitatively such striking features as vertical and largely incoherent "waterfall", its doping dependence. The controversy among the experimental groups should be also mentioned [47]. We have proposed the scenario where the whole HEA is explained by assuming coupling of single electron excitations to the MIR “boson” which, as we have stated before, is responsible also for the bond-stretching phonon anomalies. Our simplified model has allowed to describe a large number of features in agreement with those reported in experiments on HEA [19]. In our scenario, the incoherent nature of MIR plays a crucial role. In particular, it is directly responsible for the verticality of the dispersion in the "waterfall" region (see Fig. 5), its anomalously large linewidth and weight. Moreover, the coupling constant can be estimated from the slope of the linear dispersion preceeding the kink, e.g., [48] finds $\lambda \sim 1.2$. That sets an even sharper restriction on other parameters, e.g., the slope and linewidth at higher energies, distribution of spectral weight in the Brillouin zone, etc. The frequency and damping parameters of the oscillator determined in this way are again pointing towards the MIR excitation known from optics.

![Fig. 5. Examples of the dispersion curves for different values of the coupling constant and damping parameter of the MIR oscillator. Numerical solution shows that the interval of allowed values that would ensure a vertical dispersion is rather limited. We obtain $\Omega = 0.4 \text{ eV}$, $\Gamma = 0.47$, $\lambda = 1.3$ at the hole doping level $\delta = 0.15$. $E$ is the energy below the Fermi level and $K$ the momentum in the Brillouin zone.](image)
Fig. 6. The calculated location of the spectral density peak in the momentum-energy space for the parameters found above. To be compared to the experimental results in Fig. 4. However, at present stage we need to know more detailed characteristics, such as dispersion and momentum resolved spectral weight of the MIR “boson”.

The connections revealed by the above analysis can be summarized as follows. The low energy kink in the single particle electronic spectrum is caused by the bond-stretching phonon mode; the latter undergoes an anomalous damping and softening due to coupling to the incoherent electron density excitation with a large spectral weight located in the mid-infrared region (0.3-0.5) eV; the same MIR excitation is responsible for the high-energy anomaly starting at 0.4 eV and reaching out till 1.5 eV. Strong doping dependence and the energy span covering the whole occupied part of the band suggest strong electron correlations as the common origin of these phenomena and justify the respective methods used in our calculations. It is known that the phonon anomaly is common to different materials belonging to the metallic perovskite oxide family [40]. Is this accidental, or are there deeper reasons for such similarity? The proposal to use phonons as a probe of the electron density excitations in these materials opens new perspectives in their investigation as it seems that no targeted search in this direction has been thought of so far [43]. However, one can find experimental reports on existence of the low energy kink in some non-cuprate perovskites (e.g., colossal magnetoresistance manganates $La_{2-2x}Sr_{1+2x}Mn_2O_7$ [49]). Even less is known about the properties of the MIR charge excitation, although we could find indication of its existence in the material with largest phonon softening, which, at the same time, has a rather high $T_c$ too, $Ba_{1-x}K_xBiO_3$ [50]. Thus, our analysis provides sufficient arguments for the existence of possible connection between different anomalies in these materials, which has not been explored before.

References

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