Electronic correlations in iron-pnictide superconductors and beyond: lessons learned from optics

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Abstract. The Coulomb repulsion, impeding electrons’ motion, has an important impact on the charge dynamics. It mainly causes a reduction in the effective metallic Drude weight (proportional to the so-called optical kinetic energy), encountered in the optical conductivity, with respect to the expectation within the nearly free electron limit (defining the so-called band kinetic energy), as evinced from band-structure calculations. In principle, the ratio between the optical and band kinetic energies allows one to define the degree of electronic correlations. Through spectral weight arguments on the excitation spectrum, we provide an experimental tool, free from any theoretical- or band-structure-based assumptions, to estimate the degree of electronic correlations in several systems. We first address the novel iron-pnictide superconductors, which serve to set the stage for our approach. We then revisit a large variety of materials, ranging from superconductors, to Kondo-like systems as well as materials close to the Mott-insulating state. For comparison, we also tackle materials where the electron–phonon coupling dominates. We establish a direct relationship between the strength of interaction and the resulting reduction in optical kinetic energy of the itinerant charge carriers.
1. Introduction

Since the discovery of superconductivity in the iron-pnictide materials [1]–[4], the issue about the strength of their electronic correlations (i.e. the repulsive interactions among electrons) has been widely debated and is still a topic of intense investigations. This issue is of stringent importance for several families of (high-temperature) superconductors, because superconductivity transition temperatures beyond 30 K cannot be yielded within the conventional mechanism based on electron-phonon coupling [5]. In this respect, ample discussion was also generated on whether the novel family of iron-pnictides does share common features and similarities with the high-temperature superconducting cuprates (HTC), discovered more than 20 years ago [6]. In the cuprates, electronic correlations are so strong that the parent compounds, out of which superconductivity originates, are Mott insulator. On the other hand, there is mounting evidence that the parent compounds of the iron-pnictide superconductors are bad metals, where electronic correlations are sufficiently strong to place them close to the boundary between itinerancy and interaction-induced electronic localizations [7]. Moreover, in both families, superconductivity develops when magnetism, characterizing in part their phase diagram, is destroyed by doping. This led to the conjecture that exchange of magnetic fluctuations may provide the glue, binding electrons into Cooper pairs [8].

The degree of electronic correlations has a direct impact on the charge dynamics, generally evinced from the optical conductivity [7]. Thanks to the development of appropriate optical methods, this latter quantity can be achieved nowadays with great precision over an extremely broad spectral range, a prerequisite of paramount importance for tracking the implications of electronic correlations. Since electronic correlations significantly impede the mobility of the electrons, they consequently lead to a substantial reduction of the kinetic energy of the itinerant charge carriers with respect to the expectation for nearly free or non-interacting particles. Precisely, one has to distinguish among two cases; namely, quenching the Drude weight either by local (Hubbard-like) correlations or by interactions with a bosonic mode (i.e. phonons or spin fluctuations) [9]. The sum rule for the first case would reveal a spectral weight change from \( n/m_{\text{LDA}} \) (\( m_{\text{LDA}} \) being the crude mass for the local density approximation (LDA)) to \( n/m_U \) (with \( m_U \) generally bigger than \( m_{\text{LDA}} \)). For the second case, the sum rule would still lead to a total weight given by \( n/m_{\text{LDA}} \), the latter being then redistributed between a Drude peak with weight...
proportional to $n/m^*$ ($m^*$ is the renormalized mass by the bosonic mode and is greater than $m_{\text{LDA}}$) and a so-called incoherent part encountering the remaining weight.

Qazilbash et al \[10\] recently made an interesting survey of the electronic correlations by looking to a wealth of materials, ranging from conventional metals to Mott insulators. Along the same line of arguments previously introduced by Millis et al \[11\], they proposed a quantitative approach for the calculation of the ratio between the optical kinetic energy ($K_{\text{opt}}$) and the band kinetic energy ($K_{\text{band}}$). The former is obtained from the integral of the effective (Drude) metallic component of the optical conductivity (also referred to as the coherent part of the single electron excitations in an interacting metallic system), whereas the latter quantity (also known as the kinetic energy of the underlying non-interacting system) is extracted from \textit{ab initio} (tight-binding) band-structure calculations neglecting the electron–electron interactions \[7\]. The analysis proposed in \[10\] establishes a regime of moderate correlations for the iron-pnictide superconductors and leads, for a large variety of materials, to a nice correspondence between their degree of electronic correlations and the reduction of the empirical kinetic energy of the charge carriers compared with theory. Materials for which the electron–phonon coupling predominately shapes the intrinsic physical properties do not show, on the other hand, a significant reduction of the electrons’ kinetic energy (figure 3 in \[10\]).

Motivated by the work reported in \[10\], we undertook a systematic optical investigation of the Co-doped BaFe$_2$As$_2$ family, spanning the entire phase diagram \[12\]. We proposed to establish the ratio $K_{\text{opt}}/K_{\text{band}}$ exclusively from spectral weight arguments based on the experimental findings. The astonishing good correspondence of the degree of electronic correlations with the evolution of the superconducting phase in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ (i.e. the superconducting dome defined by the $T_c$ values in the phase diagram (\[13\] and references therein)) makes us confident that one could in principle expand our procedure for other materials. The goal of this paper is to review our estimation of the electronic correlations across the phase diagram of the Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ compounds and place it in a broader context by revisiting, from a similar perspective, other variably correlated materials. We will compare the iron-pnictides to other superconductors, such as HTCs, boron carbides and fullerenes, and to strongly correlated $f$- and $d$-electron systems (Kondo and heavy electron (HE) materials), as well as linear chain organic Bechgaard salts. Our review will be complemented by representative materials characterized by a charge-density-wave (CDW) ground state for which the electron–phonon coupling should dominate. Apart from the data on HTC, we otherwise made use of our own results being directly accessible. We choose them to be representative of a large variety of families on which various groups obtained equivalent results (when appropriate we will compare data from different sources), thus implying the general applicability of our method.

We show that a dedicated spectral-weight analysis can indeed reliably reveal the degree of electronic correlations and thus provide a valuable and exclusive experimental instrument, free from any \textit{ad hoc} theoretical constraints, to discriminate between various systems with different levels of correlation. We analyze the real part ($\sigma_1(\omega)$) of the optical conductivity, which is mainly obtained by first measuring the reflectivity ($R(\omega)$) over an extremely broad spectral range, extending from the far-infrared (FIR) up to the ultraviolet (UV). For some selected systems presented here, optical data were collected in the THz and microwave regions, mainly with resonant cavity techniques. $R(\omega)$ data over a wide energy interval permit us to perform reliable Kramers–Kronig (KK) transformation from which we calculate the phase of the complex reflectance and then all optical functions, including $\sigma_1(\omega)$ \[14, 15\]. The reliability of
Figure 1. The real part $\sigma_1(\omega)$ of the optical conductivity of Ba(Co$_x$Fe$_{1-x}$)$_2$As$_2$ for $x = 0, 6.1$ and $18\%$ in the FIR and the mid-infrared (MIR) spectral range at selected temperatures above and below the various phase transitions. The middle panel refers to the left $y$-axis. The inset displays $\sigma_1(\omega)$ at $300 \, \text{K}$ for $x = 18\%$, emphasizing its representative shape for all compounds at high frequencies up to the UV [12].

the KK procedure is well supported by the overall good agreement, established for all materials presented in this review, between the dc limit $\sigma_1(\omega \to 0)$ of the optical conductivity and the $\sigma_{dc}$ values extracted from the transport data.

2. Experimental results and discussion

2.1. Iron-pnictide superconductors

We start our survey by summarizing the most pertinent results on Co-doped BaFe$_2$As$_2$, which are reported in great detail in [12] and which serve as an interesting case in point. Figure 1 shows $\sigma_1(\omega)$ in the FIR and MIR energy intervals for selected Co-dopings (non-superconducting parent compound, optimal doping and non-superconducting overdoped compound) as a function of temperature. The overall temperature dependence of $\sigma_1(\omega)$ for all compounds generally agrees with data collected from other groups [16]–[26]. The resulting $\sigma_1(\omega \to 0)$ limits at $T > T_{\text{SDW}}$ or $T_c$ are in quite good agreement with the dc results, thus confirming the consistency of the KK procedure. In the parent ($x = 0$) and underdoped non-superconducting compound ($x = 0.025$, not shown [12]), $\sigma_1(\omega)$ slightly decreases below $2000 \, \text{cm}^{-1}$ in the normal state, leading to a shallow minimum prior to the onset of the (narrow) Drude component in FIR. This is the consequence of the overdamped behavior of $R(\omega)$, which in our samples seems to be more pronounced than in a few other works [16, 17, 19]–[22]. This probably originates from the somehow smaller dc conductivity of our specimens [13] than in other samples. These deviations turn out, however, to be uninfluential for the data analysis and their overall interpretation, as discussed here. The parent compound ($x = 0$) clearly displays the opening of the SDW-pseudogap at $T_{\text{SDW}} \sim 135 \, \text{K}$, while the optimally doped one ($x = 6.1\%$) gives clear-cut evidence for the development of the superconducting gap below $T_c \sim 23 \, \text{K}$. Finally, the compound ($x = 18\%$) at the opposite end of the superconducting dome is in the metallic...
Figure 2. Phase diagram of Ba(Co$_{x}$Fe$_{1-x}$)$_2$As$_2$, reproduced from [13] (left y-axis), and the average of the ratio $K_{\text{band}}/K_{\text{opt}}$ calculated at high and low temperature (right y-axis) from the Drude–Lorentz (LD) fit parameters (equation (1)) and from the spectral weight (SW) analysis with $\omega_{\text{opt}} = 500$ cm$^{-1}$ and $\omega_{\text{band}} = 2000$ cm$^{-1}$ (equation (2)). $T_c$, $T_{\text{SDW}}$ and $T_{\text{TO}}$ are the critical temperatures for the superconducting and SDW phase transition as well as for the tetragonal–orthorhombic structural transition, respectively [13]. All data-interpolations are spline lines as guide to the eyes. The inset displays the optical conductivity at 120 K for the parent compound ($x = 0$) with the total Drude–Lorentz fit and its components; i.e. the narrow and broad Drude terms, the MIR h.o. as well as the temperature-independent h.o.’s for the high-frequency interband transitions. The shaded areas emphasize the spectral weights of both Drude terms and MIR band [12]. The dashed and dashed-dotted arrows mark the cut-off frequencies $\omega_{\text{opt}}$ and $\omega_{\text{band}}$ (see text).

phase at all temperatures. Overall, there are three energy intervals characterizing $\sigma_1(\omega)$ for all Co-dopings: the effective metallic contribution at low frequencies, a rather flat MIR region covering the energy interval between 500 and 1500 cm$^{-1}$ (from now on called the MIR band) and the electronic interband transitions with onset at about 2000 cm$^{-1}$ and peaked at 5000 cm$^{-1}$ (inset, figure 1). The metallic part as well as the MIR band turn out to experience the strongest temperature dependence at $T_c$ and/or $T_{\text{SDW}}$, while the high-frequency excitations are temperature independent [12].

To account for the various contributions to the excitation spectrum, we applied the well-established phenomenological Drude–Lorentz approach [14, 15]. Besides high-frequency and temperature-independent Lorentz harmonic oscillators (h.o.) for the interband transitions with onset at $\omega \sim 2000$ cm$^{-1}$ (inset of figure 1), $\sigma_1(\omega)$ can be reproduced in great detail by adding two Drude terms for the effective metallic contribution and a broad h.o. for the MIR energy interval (inset of figure 2) [12, 20, 21, 23]. These latter phenomenological components fully describe the temperature dependence of $\sigma_1(\omega)$ at $\omega \leq 2000$ cm$^{-1}$ for all dopings ($x$). Consistent
with [20] and [23], it turns out that one Drude term is rather narrow, while the second broad one acts as a background to the optical conductivity. The two (narrow \((N)\) and broad \((B)\)) Drude terms imply the existence of two electronic subsystems. High-resolution angle-resolved photoemission (ARPES) studies recently revealed the typical electronic structure of the \(\text{BaFe}_2\text{As}_2\) family, which consists of two electron bands and two hole bands near the \(X\)-point \((\pi, \pi)\) as well as of three nearly degenerate hole bands at the \(\Gamma\) point of the Brillouin zone [27, 28]. Among the bands at the \(X\)-point, only the electron bands cross the Fermi level, forming two electron pockets around \(X\), whereas the hole bands approach but never reach the Fermi level. The outcome from ARPES matches fairly well with the prediction of LDA calculations [29]–[31]. In passing, we should however remark that the Drude–Lorentz approach is too phenomenological to allow speculations on whether the two Drude components have specific orbital character. Furthermore, it should be pointed out that with increasing doping and particularly for \(x \geq 0.061\) (for details see [12]) the broad Drude term largely dominates over the narrow one [22]. For the present discussion, the relevant quantity shaping \(\sigma_1(\omega)\) is the total spectral weight encountered in the effective metallic contribution to \(\sigma_1(\omega)\) and as such is not affected by the details of its phenomenological description.

We thus proposed a scenario where the conduction band derives from \(d\)-states and splits into two parts: a purely itinerant one close to the Fermi level and represented by the two Drude components as well as by a bottom part with states below the mobility edge and thus rather localized [12]. The latter part gives rise to the MIR band in \(\sigma_1(\omega)\), which turns out to be strongly affected by the opening of the SDW gap. The MIR band acquires weight below \(T_{SDW}\) and may be thus associated with a magnetic peak, which is due to antiferromagnetically ordered magnetic stripes [32, 33], as predicted by density functional theory ([34, 35]; [36] and references therein).

To shed light on the relative distribution of spectral weight among the various (metallic and MIR) components of \(\sigma_1(\omega)\), we have suggested to further exploit our phenomenological Drude–Lorentz description by calculating the spectral weight ratio:

\[
K_{\text{opt}} / K_{\text{band}} = \frac{\omega_p^2}{\omega_p^2 + (\omega_{\text{pMIR}}^2)^2},
\]

where \(\omega_p^2 = \sum_i \omega_{pi}^2\), \(\omega_{pi}\) are the plasma frequencies of the Drude terms, shaping the effective metallic part of \(\sigma_1(\omega)\), and \((\omega_{\text{pMIR}}^2)^2 = \sum_j S_j^2\), \(S_j^2\) is the mode strength of the \(j\)-Lorentz h.o.s contributing to the MIR band in \(\sigma_1(\omega)\) (inset of figure 2). Equation (1) thus represents the ratio between the spectral weight encountered in \(\sigma_1(\omega)\) in the Drude components \(K_{\text{opt}}\) and the total spectral weight collected in \(\sigma_1(\omega)\) up to the onset of the electronic interband transitions (i.e. Drude components together with the MIR absorption feature, \(K_{\text{band}}\)), which are the shaded areas in the inset of figure 2, respectively [12]. The Drude–Lorentz phenomenological approach allows us to disentangle the contribution due to the coherent and incoherent parts of \(\sigma_1(\omega)\), leading to a corresponding spectral weight redistribution, then reflected by the ratio \(K_{\text{opt}} / K_{\text{band}}\). We shall emphasize at this point that a Drude–Lorentz fit with appropriate components may be applied to all investigated materials in this survey. Furthermore, the fit constraints were such that the measured reflectivity and the real part of the optical conductivity were simultaneously reproduced by the identical set of fit parameters. This restrains the degree of freedom in the choice of the fit components. Details pertaining to the fits for each material presented here can be evinced from the quoted references.
Alternatively, one can extract both $K_{\text{opt}}$ and $K_{\text{band}}$ from a spectral weight analysis, directly performed on the experimental $\sigma_1(\omega)$ [10]:

$$\frac{K_{\text{opt}}}{K_{\text{band}}} = \frac{\int_{\omega_{\text{opt}}}^{\omega_{\text{band}}} \sigma_1(\omega) \, d\omega}{\int_{\omega_{\text{opt}}}^{\omega_{\text{band}}} \sigma_1(\omega) \, d\omega},$$

(2)

where $\omega_{\text{opt}}$ and $\omega_{\text{band}}$ are appropriate cut-off frequencies (inset of figure 2). Although less precise than the procedure based on the Drude–Lorentz fit (equation (1)), equation (2) allows us to achieve a model-independent estimation of $K_{\text{opt}}/K_{\text{band}}$. The prerequisite is an appropriate choice of the cut-off frequencies $\omega_{\text{opt}}$ and $\omega_{\text{band}}$. $\omega_{\text{opt}}$ defines the frequency interval of the effective metallic component to $\sigma_1(\omega)$, generally reproduced by a simple Drude term or by a combination of several Drude components within a multiband scenario. Unless stated otherwise, $\omega_{\text{opt}}$ for the materials reviewed here is located within the shallow minimum between the effective metallic part and the MIR band, generally characterizing $\sigma_1(\omega)$, and basically indicates the onset of the MIR absorption. It thus identifies the upper frequency limit of the coherent part and marks the point on the frequency axis where the Drude term tends to die and the MIR band is of comparable intensity (inset of figure 2). On the other hand, $\omega_{\text{band}}$, located at the high-frequency end of the h.o. for the MIR feature, denotes the upper limit of the incoherent part in $\sigma_1(\omega)$ and sets the onset frequency of the electronic interband transitions. It is usually identified as the zero-crossing energy of the imaginary part of the complex optical conductivity [10]. While the choice of $\omega_{\text{opt}}$ and $\omega_{\text{band}}$ is exclusively driven by the shape of the optical conductivity, the Drude–Lorentz fit may be then used in order to support and independently justify their location on the frequency axis. Thus, the denominator of equation (2) encounters the total spectral weight from both the coherent metallic (i.e. the numerator of equation (2)) and the incoherent contributions to optical conductivity. We will show that $K_{\text{opt}}/K_{\text{band}}$ after equation (1), when exploiting the fit parameters, is consistent with its estimation from the direct integration of $\sigma_1(\omega)$ (i.e. equation (2)).

By assuming the conservation of the total charge carrier density, equations (1) and (2) are in principle proportional to the ratio between the effective mass at low-energy scales, which includes all renormalizations of local character or due to spin fluctuations, and at high energies, which only includes local effects of Hubbard type [37]. Therefore, the reduction in $K_{\text{opt}}$ with respect to $K_{\text{band}}$ would solely derive from the effective mass renormalization. Obviously in a multiband scenario as applies in the iron-pnictides, it could well be that the rearrangement of the total number of charge carriers between hole and electron bands is such that the total charge carriers density is not conserved. In this case, the spectral weight reduction in the effective metallic contribution of $\sigma_1(\omega)$ might not be entirely due to the effective mass (i.e. correlation effects) [38], an issue that is very much controversial [39]–[41]. One should then carefully address the role of electron–electron interactions in both effective mass and charge carriers concentration. This is beyond the scope of the present discussion. Furthermore, we are aware of the fact that the MIR band may be contaminated by coherent contributions originating from transitions between bands close to the Fermi level (in the case of the iron-pnictides of hole–hole type) or generally by the interband transitions, located at energies higher than the MIR feature. We believe that these contributions would just add a little amount of spectral weight, essentially encountered in the low-frequency tails of their absorption features in $\sigma_1(\omega)$, into the range dominated by the MIR band, thus having a moderate impact on our estimation of $K_{\text{opt}}/K_{\text{band}}$.

As already emphasized above, our definition of $K_{\text{band}}$ may be equivalent to the LDA expectation in the case when local Hubbard-like effects are negligible. The latter assumption
is widely debated, because the size of the Hubbard $U$ governs the amount of spectral weight transferred to high energies, which was found to have non-negligible implications with respect to possible violation of the $f$-sum rule in HTC ([42] and references therein; [43, 44]). The transferred weight might be obviously relevant and could occur at energy scales out of the reach of our experiments [41]. Nevertheless, we are confident that equations (1) and (2) are an alternative estimation, exclusively obtained from the experimental findings, of the ratio between the optical kinetic energy extracted by integrating $\sigma_1(\omega)$ up to the onset of the electronic interband transitions [10] and the band kinetic energy extracted from the band structure within the tight-binding approach.

We have calculated $K_{\text{opt}}/K_{\text{band}}$ after equation (1) at high and low temperatures, yet still corresponding to the normal or SDW state\(^1\). $K_{\text{opt}}/K_{\text{band}}$ results from the sum of the (narrow and broad) Drude weights (i.e. $\omega_{p}^2 = \omega_{p,n}^2 + \omega_{p,B}^2$, the numerator in equation (1)) and the total h.o. strength of the MIR band (i.e. $(\omega_{p,\text{MIR}})^2$), as established from the fits. Our $K_{\text{opt}}/K_{\text{band}}$ for $x = 0$ is consistent with the value reported in figure 3 of [10] for the same compound, therefore reinforcing the validity of our approach. The inverse of $K_{\text{opt}}/K_{\text{band}}$, which then defines the degree of electronic correlations, is plotted in figure 2 as the average of the $K_{\text{band}}/K_{\text{opt}}$ values at low and high temperatures (see footnote 1) within the phase diagram of the Co-doped 122 iron-pnictides [13].

Setting $\omega_{\text{opt}} = 500 \text{ cm}^{-1}$ and $\omega_{\text{band}} = 2000 \text{ cm}^{-1}$, we can also estimate $K_{\text{opt}}$ and $K_{\text{band}}$ through a direct integration (equation (2)) of the experimental $\sigma_1(\omega)$ up to the respective cut-off frequencies. The chosen cut-off frequencies fully satisfy our criteria, because at $\omega_{\text{opt}}$ the Drude term and MIR h.o. are of comparable intensity and $\omega_{\text{band}}$ is the crossover energy between the MIR feature and the onset of interband transitions (inset of figure 2). Figure 2 then reproduces the average of $K_{\text{band}}/K_{\text{opt}}$ values after equation (2) at the same low and high temperatures as before. It is rather rewarding that the two approaches, based on both Drude–Lorentz fits or integrated $\sigma_1(\omega)$, lead to comparable estimations of $K_{\text{band}}/K_{\text{opt}}$ for all Co-dopings. We emphasize that a lower choice of $\omega_{\text{opt}}$ would too strongly underestimate the weight of the effective metallic components. Similarly, for a higher choice of $\omega_{\text{band}}$ the contribution of electronic interband transitions to $\sigma_1(\omega)$ would become non-negligible, thus overestimating $K_{\text{band}}$. A much lower choice of $\omega_{\text{band}}$ or a much higher choice of $\omega_{\text{opt}}$ would be unrealistic, since on the one hand the electronic interband transitions would contaminate $\sigma_1(\omega)$ at anomalously low energy scales and on the other hand the MIR feature would be underestimated. The error bar in figure 2 emphasizes the expected variation in the average value of $K_{\text{band}}/K_{\text{opt}}$ at high and low temperatures with $\omega_{\text{opt}} = 300 \text{ cm}^{-1}$ after equation (2) with respect to the ideal situation for $\omega_{\text{opt}} = 500 \text{ cm}^{-1}$ ($\omega_{\text{band}} = 2000 \text{ cm}^{-1}$ in

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1 That superconductivity in the iron-pnictides emerges in the close proximity of an antiferromagnetic (AF) ground state implies the important role of AF fluctuations in the pairing mechanism of all itinerant charge carriers. Both Drude terms are affected by the superconducting transition [23] and it is consequently legitimate to consider the total spectral weight of both of them (i.e. involving all ungapped charge carriers) for the definition of the optical kinetic energy. For low Co-dopings (particularly for $x \simeq 0.025$), the SDW instability and the resulting Fermi surface gapping at low temperatures within the SDW state obviously lead to a reshuffling of spectral weight, thus affecting both quantities $K_{\text{opt}}$ and $K_{\text{band}}$ in equation (1). Therefore, $K_{\text{opt}}/K_{\text{band}}$ after equation (1) for the underdoped compounds ($x \lesssim 0.025$) is smaller at $T < T_{\text{SDW}}$ than above. Nevertheless, it turns out that $K_{\text{band}}/K_{\text{opt}}$ both at high and low temperatures behaves similarly as a function of Co-doping, so that both estimations converge with increasing doping content and are basically identical for $x \gtrsim 0.11$. Thus, the proposed average value in figure 2 suitably gives an overall trend for the degree of electronic correlations.
Figure 3. The real part $\sigma_1(\omega)$ of the optical conductivity at selected temperatures for the linear chain Bechgaard salt (TMTSF)$_2$PF$_6$ [50, 51], the HE CeAl$_3$ [56], the ferromagnetic metal Eu$_{1-x}$Ca$_x$B$_6$ for $x = 0, 0.3$ and $0.8$ [63, 65], the Kondo insulator FeSi [58], the CDW rare-earth triellurides ErTe$_3$ [69] and the superconducting boron carbide YNi$_2$B$_2$C [46] and alkali-doped fullerene K$_3$C$_{60}$ [45] (for clarity $\sigma_1(\omega)$ has been multiplied here by a factor of three). Data for (TMTSF)$_2$PF$_6$ and CeAl$_3$ are shown with frequency scales, extending into the microwave spectral range. As an example, the Drude term and MIR h.o. of the phenomenological (LD) fit are shown for FeSi and YNi$_2$B$_2$C. Narrow ($T < T_K$) and broad ($T > T_K$) Drude behavior is also displayed for CeAl$_3$. The dashed and dashed-dotted arrows mark the cut-off frequencies $\omega_{\text{opt}}$ and $\omega_{\text{band}}$ (see text) for the integral of the optical conductivity leading to $K_{\text{opt}}$ and $K_{\text{band}}$ (equation (2)), respectively [7, 10].

As expected, a lower choice of $\omega_{\text{opt}}$ (under) overestimates the strength of the (Drude) MIR term in $\sigma_1(\omega)$ and leads to an artificial enhancement of $K_{\text{band}}/K_{\text{opt}}$, which turns out to be significant for $x \leq 0.025$. Nonetheless, the overall trend of decreasing $K_{\text{band}}/K_{\text{opt}}$...
with increasing Co-doping content is still preserved even in this extreme case. $K_{\text{band}}/K_{\text{opt}}$ thus tracks the evolution of the superconducting dome in Co-doped BaFe$_2$As$_2$. Interestingly enough, electronic correlations seem to be stronger for the parent compound and for Co dopings $x \leq 0.061$ than for those in the overdoped range. There is indeed evidence for a crossover from a regime of moderate correlations for $x \leq 0.061$ to a nearly free and non-interacting electron gas system for $x \geq 0.11$ [12].

2.2. Boron carbide, fullerene and high-temperature copper-oxide superconductors

With the goal to further generalize our approach towards the estimation of $K_{\text{opt}}/K_{\text{band}}$ based on the spectral weight redistribution of $\sigma_1(\omega)$, we recollect first of all the electrodynamic response of two other families of superconductors: the alkali-doped fullerenes (A$_3$C$_60$, A = K, Rb and Cs) and the boron carbides LNi$_2$B$_2$C ($L = \text{Lu}, \text{Tm}, \text{Er}, \text{Ho}, \text{Dy}$ and Y). Their electrodynamic response ([45] and references therein; [46]) in the normal state shares general features with $\sigma_1(\omega)$ of the HTCs (see below) [42] and iron-pnictides [12], as well. The normal state $\sigma_1(\omega)$ of YNi$_2$B$_2$C and K$_3$C$_60$ is reproduced in figure 3. There is evidence for a two-component picture: a Drude response up to $\omega_{\text{opt}}$, getting narrow with decreasing temperature, and a MIR band, extending up to $\omega_{\text{band}}$ and being reminiscent of a pseudogap-like excitation. Those generic features are emphasized by the Drude and h.o. fit components, shown for the boron carbide case (figure 3) [46]. Using the parameters of the Drude–Lorentz fit [45, 46] within equation (1), it turns out that only about 10% of the total weight (i.e. $K_{\text{opt}}/K_{\text{band}} \sim 0.1$), encountered in $\sigma_1(\omega)$ in the Drude component and MIR band up to the onset of the electronic interband transitions, can be ascribed to the itinerant charge carriers. Following our criteria (section 2.1), we set $\omega_{\text{opt}} = 110 \ (200)$ and $\omega_{\text{band}} = 2000 \ (5000) \ \text{cm}^{-1}$ for the direct integration of $\sigma_1(\omega)$ after equation (2) in fullerenes and boron carbides, respectively. We may appreciate that the Drude term and h.o. are indeed of comparable strength at energy coinciding with the definition of $\omega_{\text{opt}}$, whereas $\omega_{\text{band}}$ coincides with the high-frequency tail of the h.o. describing the MIR band (figure 3). Equation (2) leads to $K_{\text{opt}}/K_{\text{band}} \sim 6\% \ (7\%)$ for alkali-doped fullerenes and boron carbides, respectively, in quite good agreement with the result from equation (1). This would imply that both the superconducting fullerenes and boron carbides are strongly correlated materials at the verge of a Mott-insulating state. $K_{\text{opt}}/K_{\text{band}}$ from our phenomenological fit for both systems are summarized in our figure of merit (figure 4), allowing a first comparison with values previously achieved for the iron-pnictides (figure 2).

It is now compelling to compare the degree of electronic correlations between these latter families of superconductors and the high-temperature copper oxides. For the purpose of our discussion, we consider the partial, yet representative series of the underdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212 (UD), $T_c = 67 \ \text{K}$), the optimally doped YBa$_2$Cu$_3$O$_y$ (Y123 (OP), $T_c = 93.5 \ \text{K}$) and the overdoped Tl$_2$Sr$_2$CuO$_{6+\delta}$ (Tl2201 (OD), $T_c = 23 \ \text{K}$) compounds (a detailed list of references can be found in [42, 47]). Here, we directly integrate $\sigma_1(\omega)$ at $T > T_c$ (i.e. in the normal state) up to the onset of the electronic interband transitions (at about $\omega_{\text{band}} \sim 1 \ \text{eV}$), leading to the total spectral weight proportional to $K_{\text{band}}$ after equation (2). The effective Drude weight, which then defines our $K_{\text{opt}}$, is obtained by integrating $\sigma_1(\omega)$ up to a cut-off frequency $\omega_{\text{opt}} \sim 1000 \ \text{cm}^{-1}$, thus at the onset of the pseudogap excitation [47]. It is worth noting that for one school of thought, the Drude and MIR terms may merge into a single (extended) Drude component (i.e. with frequency-dependent scattering rate and effective mass of the itinerant charge carriers) in the overdamped regime [42]. Such a generalized Drude approach would not qualify for

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Figure 4. The ratio $K_{\text{opt}}/K_{\text{band}}$ calculated for selected materials, spanning a broad range of the degree of correlation. For the sake of clarity, the $K_{\text{opt}}/K_{\text{band}}$ values for groups of materials or systems are displayed with a vertical shift. Values of $K_{\text{opt}}/K_{\text{band}}$ reported here were obtained after equation (2) for the HTC compounds; those for all other materials correspond to the estimation after equation (1), i.e. based on a Drude–Lorentz fit of $\sigma_{1}(\omega)$ (see text).

optimally and underdoped cuprates. Thus, our straightforward spectral weight analysis is in this respect general and material independent. The ratio $K_{\text{opt}}/K_{\text{band}}$ is reported in figure 4 for the three chosen compounds and follows the trend of our arguments proposed above for the iron-pnictides and other superconductors and is even in quite good agreement with its estimation in [10] for other electron- and hole-doped HTCs, spanning the UD–OP regime. Our results, moreover, agree with a semi-theoretical approach to the electron- and hole-doped cuprates, reported in [11, 42]. It is worth noting that $\sigma_{1}(\omega)$ in the non-superconducting La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.26$) [48] is characterized by a simple Drude response without any signature of the pseudogap excitation, thus implying $K_{\text{opt}}/K_{\text{band}} \sim 1$ (equation (2)). This is fully consistent with a conventional free carriers scenario, as expected for simple metals such as Au or Al [10, 15].

$^2$ The $\sigma_{1}(\omega)$ spectrum of conventional metals, such as Au and Al, can be reproduced by a simple Drude response, the integral of which leads to values of the plasma frequency $\omega_p \sim \sqrt{n/m_e}$, totally in agreement with the theoretical expectation given by the charge carrier concentration ($n$) from chemical counting and the free electron mass ($m_e$) [15].
2.3. Low-dimensional organic Bechgaard salts

The linear chain organic Bechgaard salts [49] have been intensively investigated because of their rich phase diagram. At low temperatures, they provide an interesting arena in which to study the pressure (chemical and applied)-induced crossover between a spin-Peierls, an SDW and a superconducting state. At high temperatures one can study the dimensionality-driven evolution between a Mott-insulating and an incipient Fermi liquid state upon compressing the lattice. We have shown that the optical conductivity in the metallic state of (TMTSF)$_2$X (where X = SF$_6$, AsF$_6$ or ClO$_4$) is highly anisotropic and displays dramatic deviations from a simple Drude response [50, 51]. Figure 3 displays, as an example, $\sigma_1(\omega)$ for X = PF$_6$ at 300 and 20 K. One can appreciate the emergence of two prominent features with decreasing temperature: a narrow mode at zero frequency, with a small amount of spectral weight, and a mode centered around 200 cm$^{-1}$, with nearly all the spectral weight expected for the relevant number of carriers and single particle band mass (figure 3). We argued that these features are characteristic of a quasi-one-dimensional (1D) half- or quarter-filled band with Coulomb correlations, and ascribed the finite-energy mode to the correlation (pseudo)gap. We proposed a scenario consistent with doped Mott-semiconducting behavior of the organic Bechgaard salts [50, 51], for which a confinement–deconfinement crossover takes place with increasing chemical pressure. The interchain coupling, represented by the transverse charge transfer integral ($t_\perp$) between the chains, induces indeed a self-doping of the Mott-insulating state of the strict 1D limit (i.e. $t_\perp = 0$), thus populating the upper Hubbard band and leading to the tiny Drude weight encountered in the zero-energy resonance of $\sigma_1(\omega)$ (figure 3). The deconfinement of the charge transport was recently confirmed by optical investigation under applied pressure on (TMTTF)$_2$AsF$_6$ [52]. $K_{\text{opt}}/K_{\text{band}}$ was estimated at 20 K, where the characteristic features in $\sigma_1(\omega)$ are very much distinct. It thus corresponds here to the ratio (equation (1)) between the spectral weight of the zero-energy mode, fitted with a (narrow) Drude term, and the total weight of both the zero- and finite-energy modes (the latter mode is fitted with a Lorentz h.o.) [50]. Alternatively, we may apply equation (2) with $\omega_{\text{opt}} \sim 20$ cm$^{-1}$ and $\omega_{\text{band}} \sim 4000$ cm$^{-1}$, which after our convention (section 2.1) mark the onset of the incoherent part and interband transitions, respectively. We remark that the total weight also corresponds to the weight included in $\sigma_1(\omega)$ up to $\omega_{\text{band}}$ at 300 K. Both equation (1), exploiting the Drude–Lorentz fit, and equation (2) from the integration of the narrow Drude as well as of the finite-energy mode result in $K_{\text{opt}}/K_{\text{band}} \sim 1\%$ (figure 4), which is again consistent with the Mott limit.

2.4. Kondo systems

Another interesting class of correlated systems is the family of HE and more generally Kondo-like materials. Electron–electron interaction leads to two characteristic excitations: a renormalized Drude response and an MIR peak ([53] and references therein; [54]). The latter originates from a dynamical, correlation-induced gap, as evinced from a many-body theoretical approach based on the periodic Anderson model [55]. At low temperatures, it can be viewed as optical gap between two renormalized quasi-particle bands. Paramount examples of HEs are CeAl$_3$, CePd$_3$ and UPt$_3$, just to name a few. Figure 3 displays $\sigma_1(\omega)$ above and below the Kondo temperature $T_K \sim 3$ K for CeAl$_3$ [56], which is very much representative of the electrodynamic response of HE systems [53]. The spectrum above $T_K$ is compatible with a simple Drude response, given by the charge carriers concentration from chemical counting and the band mass.

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$m_b \sim 2m_e$, and can be used to estimate $K_{\text{band}}$. $\sigma_1(\omega)$ below $T_K$ is characterized by the narrow Drude contribution, due to the effective mass renormalization, and by a broad tail defining the MIR band (incoherent) contribution [54, 55]. The spectral weight of the renormalized (narrow) Drude response defines $K_{\text{opt}}$. The Drude terms above and below $T_K$ are explicitly shown in figure 3. Their high-frequency tails, where they both merge into the (incoherent) broad MIR band and interband transitions, allow us to set $\omega_{\text{opt}} \sim 6 \text{ cm}^{-1}$ and $\omega_{\text{band}} \sim 4000 \text{ cm}^{-1}$, respectively. $K_{\text{opt}}/K_{\text{band}}$ at $T < T_K$ in prototype HEs from both equations (1) and (2) oscillates between 0.003 and 0.02; that is, only about 0.3–2% of the expected total spectral weight in the nearly free electron or simple band-metal limit is effectively encountered in the (narrow) Drude response of $\sigma_1(\omega)$ [53]–[56]. These values of $K_{\text{opt}}/K_{\text{band}}$ also place the HE systems within the strongly correlated regime, similar to the situation observed in the low-dimensional organic systems (figure 4). It is worth noting that our experimental estimation of $K_{\text{opt}}/K_{\text{band}}$ is particularly suitable in this context. In fact, it is not plagued by the annoying role of the localized $f$-electrons, which would make $K_{\text{band}}$ from band-structure calculations less trustable and poorly reliable.

Among the highly correlated electron systems, various rare-earth compounds known as hybridization-gap semiconductors or Kondo insulators [53, 57] have also attracted considerable interest recently. The cubic compound FeSi is a prominent example, particularly suitable for investigating aspects of the electronic properties of a $d$-transition metal system that might be related to features of correlation effects in $f$-electron materials. At low frequencies, Anderson–Mott localization behavior was established from the low-temperature optical conductivity, while at high frequencies the excitation spectrum resembles that of a conventional semiconductor [58]. Figure 3 displays $\sigma_1(\omega)$ of FeSi at 300 K, which is characterized by a metallic contribution below $\omega_{\text{opt}} \sim 500 \text{ cm}^{-1}$ and by a broad bump peaked at about 700 cm$^{-1}$ and extending up to $\omega_{\text{band}} \sim 1500 \text{ cm}^{-1}$ [58]–[62]. The latter feature is an incipient hybridization-induced pseudogap, which evolves in the semiconducting gap excitation at low temperatures (e.g. at 10 K, figure 3)$^3$. A single Drude term and one h.o. (figure 3) fully describe $\sigma_1(\omega)$ at 300 K below $\omega_{\text{band}}$. We may again remark that, according to our criteria (section 2.1), $\omega_{\text{opt}}$ located at the minimum within the depletion of $\sigma_1(\omega)$ between its metallic and MIR band features is close to the energy where the Drude term and h.o. have equal strength. At $\omega_{\text{band}}$, defined as the zero-crossing energy of the imaginary part of the complex optical conductivity, the MIR band rapidly vanishes and the interband transitions acquire considerable weight. We did not observe any major difference between the estimations of $K_{\text{opt}}/K_{\text{band}}$ after equation (1) with the Drude–Lorentz fit parameters [58, 61, 62] or through direct integration of $\sigma_1(\omega)$ (equation (2)). The ratio of the spectral weight belonging to the metallic state up to $\omega_{\text{opt}}$ with respect to the total one up to $\omega_{\text{band}}$, including the semiconducting MIR gap, leads to $K_{\text{opt}}/K_{\text{band}}$ at 300 K of about 0.4 (figure 4), thus placing FeSi in the regime of moderate electronic correlations.

2.5. Ferromagnet europium hexaborides

Materials exhibiting colossal magnetoresistive effect are of high current interest in the field of solid-state physics, primarily because of their potential technological applications. The well-known manganites and Ca-doped EuB$_6$, for which the onset of ferromagnetism is accompanied

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$^3$ The sharp lines in FeSi below $\omega_{\text{opt}}$ at 10 K (figure 3) are optical phonon modes [58, 59].
by a dramatic reduction in the electrical resistivity, have been intensively studied. By magneto-optical investigations of the Eu$_{1-x}$Ca$_x$B$_6$ series [63, 64], we revealed a phase diagram in support of a scenario based on the double-exchange model ([65] and references therein). That model foresees the close proximity of the Fermi level and a magnetization-dependent mobility edge so that a ferromagnetic metal–(Anderson) insulator transition (at $T = 0$) occurs on increasing the Ca content above a critical Ca concentration ($x_{\text{MI}}$) of about 0.5. The Ca doping induces a drift of the mobility edge such that above $x_{\text{MI}}$ it goes past the Fermi energy and the spin polarization due to the ferromagnetic transition no longer releases any of the localized states. The Drude weight is thus insensitive to the spin polarization above $x_{\text{MI}}$ [65]. The fingerprint of such a metal–insulator crossover (at $T > 0$) was tracked by the spectral weight changes in the Drude component as a function of both the magnetic field and the temperature [65]. Here, we wish to show that the ferromagnetic metal–insulator transition upon Ca doping is tightly related to the degree of electronic correlations. For this purpose, we look at the optical conductivity $\sigma_1(\omega)$ above the ferromagnetic phase transition ($T_C$) for selected Ca doping, shown in figure 3. It gives evidence for a metallic component up to $\omega_{\text{opt}} \sim 300$ cm$^{-1}$, with a broad high-frequency tail, extending up to $\omega_{\text{band}} \sim 2000$ cm$^{-1}$ and due to localized charge carriers. We can calculate $K_{\text{opt}}/K_{\text{band}}$ for selected Ca doping at $T > T_C$ from the Drude–Lorentz fit parameters [65] after equation (1) or alternatively within the spectral weight arguments of equation (2) by considering the ratio between the effective metallic (Drude) spectral weight (i.e. up to the cut-off energy $\omega_{\text{opt}}$) and the overall one encountered in $\sigma_1(\omega)$ below $\omega_{\text{band}}$, thus including the excitations due to the charge carriers located below the mobility edge. Both ways give almost identical results. Figure 4 shows $K_{\text{opt}}/K_{\text{band}}$ for $x = 0.3$ (i.e. $x \leq x_{\text{MI}}$) and $x = 0.8$ (i.e. $x \geq x_{\text{MI}}$). Eu$_{1-x}$Ca$_x$B$_6$ with Ca doping $x < x_{\text{MI}}$ appear to fall in the regime of moderate electronic correlations, while for doping approaching or above $x_{\text{MI}}$ there is evidence of stronger ones. The ferromagnetic metal–insulator transition, observed at low temperatures on Ca doping, thus seems to go hand in hand with the electronic correlations’ crossover, estimated through our spectral weight arguments.

2.6. Charge-density-wave (CDW) materials

We conclude our survey by addressing the broken symmetry ground state due to the formation of a CDW condensate, which derives from the Peierls transition [66]. The paradigm of CDW-forming materials is the quasi-1D compounds, but electronically driven CDW states were also found in novel 2D-layered compounds [67]. The physics of low-dimensional CDW systems recently experienced a revival of interest. In particular, 2D CDW materials were thoroughly re-investigated, an effort motivated in part by the fact that high-temperature superconductivity in the cuprates may emerge from a peculiar charge ordering in 2D through the tuning of relevant parameters (see section V in [68]). In figure 3, we display $\sigma_1(\omega)$ for a prototype-layered CDW system, ErTe$_3$, both in the normal and the CDW state [69]. Above the CDW transition temperature ($T_{\text{CDW}} \sim 265$ K)$^4$, besides the metallic contribution up to $\omega_{\text{opt}} \sim 1500$ cm$^{-1}$ there is a broad MIR high-energy tail up to $\omega_{\text{band}} \sim 5000$ cm$^{-1}$. In quasi-1D CDW materials, such as the well-known K$_{0.3}$MoO$_3$ (not shown), the broad MIR feature clearly appears as a pseudogap-like excitation [70]. The latter feature, developing above $T_{\text{CDW}}$, is ascribed to a precursor CDW gap because of fluctuation effects and evolves into the CDW single-particle excitation below $T_{\text{CDW}}$.

$^4$ ErTe$_3$ and HoTe$_3$ also display a second CDW transition at 157 and 110 K, respectively [69]. Here, we limit our discussion to the normal state and deep in the CDW states, with respect to both CDW transitions.
This can be appreciated in figure 3 for the ErTe$_3$ case, where indeed the single-particle peak due to the CDW gap clearly emerges at $T < T_{\text{CDW}}$. These optical properties can be well reproduced by appropriate Drude–Lorentz fits, as detailed in [69, 70]. $\sigma(\omega)$ in the CDW state (figure 3) further allows us to verify the consistency of our choice for $\omega_{\text{opt}}$ and $\omega_{\text{band}}$ with the criteria given in section 2.1; $\omega_{\text{opt}}$ is located in the shallow minimum between the metallic component and CDW (pseudo)gap, whereas $\omega_{\text{band}}$ coincides with the high-frequency tail of the MIR band. Therefore, $K_{\text{band}}$ results from the total spectral weight, given by the sum of the effective metallic contribution (then defining $K_{\text{opt}}$) and the weight encountered in the MIR feature due to the Fermi surface gapping as a consequence of the CDW precursor effects. For typical 1D materials, such as K$_{0.3}$MoO$_3$, the resulting $K_{\text{opt}}/K_{\text{band}}$ from both approaches (i.e. equations (1) and (2)) is astonishingly small, of the order of 0.1, whereas for the 2D rare-earth tritellurides, such as ErTe$_3$ or HoTe$_3$, it is about 0.8–1 (figure 4). This would imply that the reduction of the electrons’ kinetic energy is negligible in 2D materials, as usually expected when electron–phonon coupling is at work. That $K_{\text{opt}}/K_{\text{band}}$ is, on the other hand, substantially reduced in truly chain-like 1D systems possibly originates from an extremely strong electron–phonon coupling, leading to the formation of polarons [71]. The interesting case of 1D CDW materials allows us to broaden the notion of reduced kinetic energy not only as a consequence of electron–electron but also of electron–phonon interaction, thus generalizing the impact of interactions on the charge dynamics as well as ultimately the concept of correlation.

3. Conclusion

The present survey over a large wealth of materials gives rather clear-cut evidence that the reduction of the (Drude) metallic spectral weight in the excitation spectrum, evinced experimentally, may provide an opportunity to establish the strength of interaction (electron–electron as well as electron–phonon). The total spectral weight encountered in the optical conductivity up to the onset of the electronic interband transitions, which is considered to be representative of the expected weight in a normal (uncorrelated) metallic state, turns out to be usually redistributed into a well-distinct Drude resonance and a so-called incoherent part, identified throughout the paper with the generic MIR-band concept. We figure out that the stronger the Coulomb repulsion or the electron–phonon coupling, the more important the reshuffling of spectral weight into the incoherent part of $\sigma(\omega)$ and the stronger the deviation of the charge dynamics from a simple band-metal response. The strongly correlated regime, eventually compatible with a Mott insulating or polaron dominated state, is thus characterized by a pronounced reduction of the effective metallic spectral weight. Consequently, we may reformulate the concept of the degree of correlation by the more general notion of degree of renormalization of the low-energy electronic states due to electron correlation and/or electron–boson interaction.

Since the origin of interactions may be very diverse, the physical meaning of the MIR feature, due to the incoherent part of $\sigma(\omega)$, is also very different from system to system. Similarly, the reduced Drude weight, as effective integrated quantity, may originate from changes of the charge density as well as of the mass enhancement of the itinerant carriers, depending on the physics involved in the investigated material. Here, we simply made the assumption that irrespective of the intrinsic driving mechanisms the effective (reduced) Drude weight should be compared to the total one encountered in $\sigma(\omega)$ below the interband transitions. It remains to be seen how the total weight up to $\omega_{\text{band}}$ deviates from the LDA expectation.
in the nearly free electron limit. In other words, a systematic check of the influence of the electron interaction $U$ in an LDA + $U$ picture is highly desired. Nevertheless, we believe that our definition of the degree of correlation from spectral weight arguments in terms of the $K_{opt}/K_{band}$ ratio would be little influenced by those deviations, thus representing a truly experimental and reliable method towards its estimation.

The impossibility, within our spectral weight arguments, to disentangle between charge density and effective mass changes is of particular relevance when a multiband scenario would apply [39, 40]. Not only changes in the effective optical mass but even changes in the effective Fermi surface volume may lead to substantial renormalization of the optical Drude weight [41]. While reduction of the effective Drude weight originating from the charge density effect could also reflect the impact of correlation effects, this issue is not negligible and could eventually represent a limitation of our approach.

Another important issue that needs further theoretical insights concerns the role played by the intrinsic dimensionality of anisotropic interacting electron gas systems in shaping the spectral weight distribution. Our approach as well as alternative approaches (e.g. [10]) obviously apply along the chain direction or within the planes only, where the signatures due to correlation effects most clearly appear.

Finally, even though our survey covers a vast group of interesting materials spanning a large area of topics at present of interest in condensed matter, it would be extremely instructive to broaden our analysis to more exotic superconductors (e.g. Ba–Pb–Bi–O systems) or to systematically approach the proximity to AF phases and Mott states (like in several organics) from the perspective of the degree of electronic correlations evinced from the electrodynamic response.

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