Glue function of optimally and overdoped cuprates from inversion of the Raman spectra

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Abstract

We address the issue of identifying the mediators of effective interactions in cuprates superconductors. Specifically, we use inversion theory to analyze Raman spectra of optimally and over-doped La₂₋ₓSrₓCuO₄ samples. This allows us to extract the so-called glue function without making any a priori assumption based on any specific model. We use instead two different techniques, namely the singular value decomposition and a multi-rectangle decomposition. With both techniques we find consistent results showing that: (i) two distinct excitations are responsible for the glue function, which have completely different doping dependence. One excitation becomes weak above optimal doping, where on the contrary the other keeps (or even slightly increases) its strength; (ii) there is a marked temperature dependence on the weight and spectral distribution of these excitations, which therefore must have a somewhat critical character. It is quite natural to identify and characterize these two distinct excitations as damped antiferromagnetic spin waves and damped charge density waves, respectively. This sets the stage for a scenario in which superconductivity is concomitant and competing with a charge ordering instability.

Keywords: high temperature superconductivity, Raman spectra, glue function

(Some figures may appear in colour only in the online journal)
a compelling evidence due to the recent direct observation by resonant x-ray scattering [25–30].

The evidence of charge ordering and the simultaneous natural presence of spin waves emanating from the nearby AFM state raises the issue of coexistence, interplay, and possible causal relationship between charge and spin degrees of freedom. An early point of view is that charge ordering arises from Coulomb-frustrated charge segregation due to short-range (essentially instantaneous) magnetic interactions [31, 32] or/and due to non-magnetic (e.g. phononic [33, 34], charge-transfer [6, 35]... ...) attractive forces. In both cases, the concomitant presence of charge-ordering fluctuations ‘enslaves’ the spin degrees of freedom, allowing them to survive up to large doping. This tight entangling of spin and charge degrees of freedom is also an intrinsic feature of the ‘stripe’ concept [36]. A complementary, more recent view, is that collective retarded spin fluctuations may give rise to attraction in the particle-hole channel inducing a charge-density wave instability [37, 38]. At the moment, it is unclear and debated whether these two points of view are really distinct or just some kind of ‘egg-and-chicken’ issue arising from an underlying continuous interplay between charge and spin degrees of freedom. In any case it seems by now rather natural to assume that charge-density waves and spin-density waves coexist and it is quite interesting to investigate the relative weight of these different excitations in mediating the effective electron–electron interaction in cuprates in the different doping and temperature regimes. This is precisely the focus of the present work.

The presence of two distinct sources of glue in cuprates was found both in optical [39, 40], ARPES [41, 42] and Raman [23, 44] experiments. These experiments identify the so-called ‘glue function’ $\alpha^2 F(\omega)$, introduced by Eliashberg to characterize the spectral distribution and the strength of the electron-phonon coupling in the superconducting pairing [45]. From optical conductivity it is found that the glue function has a double structure with a rather narrow and temperature dependent peak at low frequencies (below $10^3$ cm$^{-1}$) and a broad structure extending up 3000–4000 cm$^{-1}$. From the characteristic frequency ranges it is quite natural to attribute the first peak to phononic excitations, while the mediators responsible for the broad peak should be diffusive magnetic excitations [46]. However, neutron scattering experiments show that the magnetic excitations have a rather rich spectral structure, [47, 48] which besides the broad high-frequency peak, also display a low-frequency peak. Moreover, time-resolved optical spectroscopy finds that also the low-frequency peak could be related to electronic excitations [40]. Thus the attribution and identification of the various excitations responsible for the effective electron–electron interaction is still an open relevant issue. In this framework, the analysis of Raman spectra is interesting because this technique is able to explore selectively different regions of the electronic Brillouin zone allowing to extract more detailed information on the electronic excitations than the optical conductivity. This selectivity of Raman spectra was already exploited in [23] to identify and characterize the double nature of the electronic mediators, but the analysis was carried out fitting the experiments by analytic expressions arising from a specific assumption on the form of the charge and spin collective excitations. In particular the standard diffusive form of nearly critical overdamped collective modes near gaussian quantum critical points was assumed. This not only allowed a good fitting of the spectra, but allowed to attribute the markedly different shape of the Raman spectra in the different channel (the $B_{1g}$ and $B_{2g}$ scattering channels obtained by differently polarizing the incoming and outgoing photons of the Raman scattering) to different, spin and charge, excitations, which acted differently on the spectra according to their different characteristic wavevectors. Although successful, this approach was starting from specific assumptions on the form of the mediators, which had the single spectral structure of overdamped modes. It is therefore quite important to test and deepen the conclusions of [23] with a more flexible and general approach. In this work we apply two different numerical inversion techniques to extract the glue function from the Raman spectra and we draw consistent conclusions: (i) two distinct excitations are responsible for the glue function, which have completely different doping dependence; (ii) there is a marked temperature dependence of the weight and spectral distribution of these excitations, which therefore must have a somewhat critical character. Indeed, if they were just the result of low-energy phonon-mediated interaction and high-energy local excitations due to strong-correlation effects, any temperature dependence would be much weaker.

Our analysis is purposely carried out on optimally and overdoped samples to highlight the specific features of the bosonic interaction mediators only. This analysis could of course be extended to the underdoped regime, where, however, the presence of a pseudogap in the fermionic spectrum introduces additional temperature, doping, and momentum dependencies, which would be superimposed to those of the bosonic mediators. This would make the interpretation of the data much less conclusive and informative.

The paper is organized as follows: in section II we describe the two numerical methods adopted to extract the glue function from the Raman spectra. Sections III and IV contain the results of our analysis of the glue functions and of their low-frequency behavior. Finally, section V reports and discusses our concluding remarks. Appendices A and B contain technical details on the implementation of the inversion procedure, while appendix C contains details on the extraction of the low-frequency properties of the glue function.

II. Glue function and Raman spectra

In this work we analyze Raman spectra from [23] in the $B_{1g}$ and $B_{2g}$ channels in $La_{2-x}Sr_xCuO_4$ (LSCO) samples for different temperatures and doping levels. Electronic Raman scattering is a bulk (nearly surface-insensitive) probe and it measures a response function $\chi(z)$ analogous to that of the optical conductivity. The Raman response function can be expressed [52] as a function of the so-called memory function $M(\zeta)$ as $\chi(z) = \chi_0 M(z)/(z + M(z))$ for complex arguments $z$, where $\chi_0$ is the (real) ‘bare’ response function computed in absence of any scattering process. The imaginary part of the response function, $\chi''$, can be written as:
\[
\chi''(\omega) = \frac{\omega \mathcal{M}''(\omega)}{\mathcal{M}'(\omega) + \omega^2 + [\mathcal{M}'(\omega)]^2}.
\] (1)

Here, the real and imaginary parts of the memory functions, \( \mathcal{M}'(\omega) = \mathcal{M}'(\omega) + i\mathcal{M}''(\omega) \), are function of the real frequency \( \omega \), and they are related by a Kramers–Kronig (KK) transformation. In turn, \( \mathcal{M}''(\omega) \) can be expressed in terms of the glue function \( \alpha^2 F(z) \) by means of the integral expression

\[
\mathcal{M}''(\omega) = \int_0^\infty dz \, K(\omega, z) \alpha^2 F(z),
\] (2)

with the kernel

\[
K(\omega, z) = \frac{2 \omega \coth \left( \frac{z}{2T} \right) - (z + \omega) \coth \left( \frac{z + \omega}{2T} \right)}{z - \omega}.
\] (3)

Therefore, \( \chi'' \) is a non linear functional of \( \alpha^2 F \). The extraction of \( \alpha^2 F \) from experimental data is possible by approximating it as a linear combination of suitable basis functions as

\[
\alpha^2 F(\omega) \approx \sum_{\alpha=1}^{N} c_\alpha \phi_\alpha(\omega).
\]

Once a basis is chosen, one may optimize the coefficients \( c_\alpha \) so as to fit the experimental data. In this way, equation (2) can be rewritten as

\[
\mathcal{M}''(\omega) = \sum_{\alpha} c_\alpha A''_\alpha(\omega),
\]

where \( A''_\alpha(\omega) = \int_0^\infty dz \, K(\omega, z) \phi_\alpha(z) \), so that equation (1) becomes

\[
\chi''(\omega, \{c_\alpha\}) = \frac{\sum c_\alpha \omega A''_\alpha(\omega)}{\left[ \sum c_\alpha A'_\alpha(\omega) + \omega^2 + [\sum c_\alpha A''_\alpha(\omega)]^2 \right]},
\] (4)

where \( A_\alpha''(\omega) \) is the KK transform of \( A_\alpha'(\omega) \). The coefficients \( c_\alpha \) are then used as fit parameters. We used and compare the results obtained with two very different strategies for the basis functions, the first one based on a multi-rectangle decomposition of the glue function, and the second based on the singular vector decomposition of the kernel \( K \).

- **Multi-rectangle decomposition (MRD).** The glue function is approximated by a piecewise constant function, which corresponds to choosing a partition of the frequency axis \( \omega_1 < \ldots < \omega_N < \omega_{N+1} \) and \( N \) non-overlapping box functions \( \phi_\alpha(\omega) = 1 \) for \( \omega_1 < \omega < \omega_{\alpha+1} \) and \( \phi_\alpha(\omega) = 0 \) elsewhere, as the basis functions. The fitting parameters \( c_\alpha \) are the heights of the box functions. In the MRD approach the functions \( A''_\alpha(\omega) \) can then be computed analytically (see appendix A for further details).

- **Singular Vector Decomposition (SVD).** The starting point of this approach is the expansion of the integral kernel equation (2) as a sum of diadic operators:

\[
K(\omega, z) = \sum_{\alpha=1}^{\infty} \sigma_\alpha \psi_\alpha(\omega) \phi_\alpha(z)
\]

where the singular values \( \sigma_\alpha \) are nonnegative and in decreasing order, and \( \{\psi_\alpha\} \) and \( \{\phi_\alpha\} \) are sets of orthogonal functions, called the left and right singular vectors, respectively. In this case we approximate the glue function using as basis the first \( N \) right-singular vectors of the singular vector decomposition of the kernel [53, 54]. This allows us to define \( A''_\alpha(\omega) = \sigma_\alpha \psi_\alpha(\omega) \). Notice that the integral kernel of our physical problem has to be properly regularized before the expansion thus leading to slightly different expressions; we refer to the appendix B for a detailed derivation.

In both cases coefficients \( c_\alpha \) are computed by minimizing the square distance \( \Delta^2 \) between the theoretical response function and the experimental one. In order to do this, the functions \( A' \) and \( A'' \) are computed at the experimental points \( \omega_i, i = 1, \ldots, N_{\exp} \). We then look for

\[
\min_{\{c_\alpha\}} \Delta^2(\{c_\alpha\}) = \sum_{i=1}^{N_{\exp}} (\chi''_{\exp, i} - \chi''_{\text{th}, \{c_\alpha\}}(\omega_i))^2,
\] (5)

where \( \chi''_{\text{th}, \{c_\alpha\}}(\omega_i, \{c_\alpha\}) \) is computed according to equation (4), and \( \chi''_{\exp, i} \equiv \chi''_{\exp}(\omega_i) \) are the measured values. The minimization is constrained by the requirement \( \alpha^2 F(\omega) \geq 0 \), which can be written in the MRD case as \( c_\alpha \geq 0 \), and in SVD case as \( \sum_\alpha c_\alpha \phi_\alpha(z) \geq 0 \), for all possible values of \( z \). Further details about the choice of the functions \( \phi_\alpha \) and \( \chi'' \), and in the discretization and in the fitting procedures can be found in appendices A (MRD) and B (SVD).

### III. B1g and B2g Glue Functions: Doping and temperature dependence

We analyze Raman spectra of La_{2-x}Sr_xCuO_4 samples focusing on three different doping levels (\( x = 0.15, 0.20, 0.25 \)) and three different temperatures (approximately 50, 100 and 200 K; the true temperatures for different doping levels are slightly different), to highlight the doping and temperature evolution. In figure 1 we show the glue functions obtained with both MRD and SVD techniques by analyzing data of three different doped samples (\( x = 0.15, 0.20, 0.25 \)) at \( T \sim 50 \) K, for both the B1g and B2g channels.

The glue function obtained by SVD has an oscillatory character, due to the fact that it is obtained by summing up oscillating functions with increasing frequency, similarly to a Fourier expansion. (See figure B1 for representative examples of the basis functions.) On the other hand, the MRD approach may yield very irregular glue functions. Some sort of indeterminacy is an inevitable consequence of the structure of the problem. Integral kernels act as low-pass filters, and therefore only the low-frequency components of the glue function, related to the largest singular values, can be reliably determined from the (noisy) data. (See figure B3 for the magnitude of the singular values of the kernel.) Instead of relying on arbitrary smoothing techniques, we proceeded in a pragmatic twofold way. On the one hand we considered integrated quantities, which are relatively insensitive to both discontinuities and oscillations in the glue functions, and therefore allow to extract robust relevant informations. On the other hand, one can notice that despite the different ‘looks’ of the glue functions obtained with the two approaches, some features are common and therefore more reliable. Indeed, both approaches yield in both channels two structures: a low-frequency peak (\( \omega \lesssim 500 \text{ cm}^{-1} \)) and a broader structure.
using the SVD (red) and MRD (black) methods, for three different doping levels ((a) and (d), $x=0.15$; (b) and (d), $x=0.20$; (c) and (f), $x=0.25$), for both $B_{1g}$ ((a)–(c)) and $B_{2g}$ ((d)–(f)) channels at low temperature ($\sim 50$ K). The insets show the experimental data (grey circles) and the response functions computed from the fitted glue functions.

Figure 1. Fitted glue functions $\alpha^2F(z)$ using the SVD (red) and MRD (black) methods, for three different doping levels ((a) and (d), $x=0.15$; (b) and (d), $x=0.20$; (c) and (f), $x=0.25$), for both $B_{1g}$ ((a)–(c)) and $B_{2g}$ ((d)–(f)) channels at low temperature ($\sim 50$ K). The insets show the experimental data (grey circles) and the response functions computed from the fitted glue functions.

in the range 500–3000 cm$^{-1}$. The $B_{1g}$ channel appears to be suppressed as the doping is increased, while the secondary structure in the $B_{2g}$ glue function shifts to lower frequencies as the doping is increased. These behaviors are also present at $T=100$ and 200 K. This latter case is reported in figure 2. Due to the behavior of the kernel at high temperature (see discussion in appendix B.4), the structures of the glue functions are rougher, but still the main features found at $T=50$ K.
IV. Low-frequency analysis: temperature and Doping dependence

The previous analysis show little temperature-dependence of the global properties of the glue function. However, the low-frequency part of the Raman response function has a clear dependence on temperature. A low-frequency expansion of the glue function allows us to study the behavior of the glue function at frequencies \( \omega \lesssim T \). As detailed in appendix C.1, the relation between the Raman response function \( \chi'' \) and the memory function \( M \) can be approximated for finite temperatures and small frequencies as

\[
\chi''(\omega) = \chi_0 \left[ \frac{\omega}{M_0''} + O(\omega^3) \right].
\]

Therefore, the low-frequency behavior of the response function is governed by the imaginary part of the memory function at zero frequency, \( M_0'' \). Interestingly, \( M_0'' \) can be directly connected to the low-frequency behavior of the glue function \( \alpha^2 F(z) \). As detailed in appendix C.2, \( M_0'' \) is proportional to an ‘effective’ slope \( s_{\text{eff}} \) of the glue function, obtained by a weighted average of \( \alpha^2 F(z) \) with a fast decaying weight, similar to \( \exp(-z/T) \). The relationship between \( s_{\text{eff}} \) and \( M_0'' \) is given by

\[
s_{\text{eff}} = \frac{3M_0''}{4\pi T^2}.
\]

If the glue function is approximately linear for \( z \lesssim T \), then \( s_{\text{eff}} \) is approximately equal to its slope. On the other hand, if the glue function has two peaks, one for \( z \lesssim T \) and the other one in \( z \gtrsim T \), the quantity \( s_{\text{eff}} \) is related to the magnitude and steepness of the first peak only, similarly to the ratio \( W_0/W_{\text{tot}} \) introduced before. As already discussed, while the derivative of the glue function cannot be reliably estimated from the fitted glued function without some kind of smoothing procedure, integrated quantities, like \( s_{\text{eff}} \) in (7), are much better suited for the analysis of the glue function properties.

We find that \( M_0'' \) displays a clear temperature trend, which is well described as a \( T^{1/2} \) scaling for both the channels. Furthermore, the two channels have a marked opposite dependence on the doping. If we use a simple linear model \( M_0'' \propto (a + bx) \) to include the effect of the doping, we obtain the following expressions:

\[
M_0''(T, x, B_{1g}) \approx (190 - 570x) \cdot \sqrt{T} \text{ cm}^{-1} \text{ K}^{-1/2},
M_0''(T, x, B_{2g}) \approx (300x) \cdot \sqrt{T} \text{ cm}^{-1} \text{ K}^{-1/2}.
\]

These scalings reflect into similar scalings for the slope of the Raman response at zero frequency and effective slopes \( s_{\text{eff}} \) of the glue functions (see appendix C and figures therein for more details). These results are in agreement with the analysis shown in the previous section. The total weight \( W_{\text{tot}} \) in the \( B_{1g} \) channel decreases almost linearly with the doping, but not \( W_{500}/W_{\text{tot}} \), therefore reducing \( s_{\text{eff}} \). On the other hand, in the \( B_{2g} \) channel, the total weight is constant, but \( W_{500}/W_{\text{tot}} \) increases with \( x \), as the low-frequency peak ‘drains’ spectral weight from the second one, the slope \( s_{\text{eff}} \) also increases.
V. Discussion and conclusions

The Raman spectra in the two, B_{1g} and B_{2g}, channels are markedly different: the B_{1g} spectra have a rounded rise at low frequency until a new rounded growth takes place above 2000 cm^{-1}, while the B_{2g} spectra have a flattish shape at low/intermediate frequencies until again a rounded growth takes place above 2000 cm^{-1}. This difference naturally stems from different physical mechanisms ruling the low/intermediate-frequency scattering mechanisms. According to the most popular scenarios of superconducting cuprates, spin and charge collective excitations are the most likely candidates as mediators of scattering. In a previous work [23] we noticed that the different direction and magnitude of the characteristic wavevectors of these excitations produce different contributions in the two channels of the Raman spectra. Specifically, it was found that charge fluctuations with characteristic wavevectors q_s \approx (\pm \pi/2, 0, 0) (in reciprocal lattice units) are more visible in the low-frequency spectra in the B_{2g} channel. On the contrary, spin fluctuations with characteristic wavevectors q_1 \approx (\pm \pi \mp \pi, \pm \pi, \pm \pi \mp \pi) (in reciprocal lattice units) contribute more in the low-frequency spectra of the B_{1g} channel. At higher frequencies both modes contribute to both channels. While the previous analysis assumed a phenomenological quantum critical form of the spin and charge excitations leading to analytic but obviously model-dependent glue functions, our main goal here is to put under scrutiny the previous results with an unbiased general analysis. Therefore within a memory-function approach we extracted the glue function directly form the Raman spectra without any assumption. To this purpose, we used two inversion methods: the SVD and the MRD. The overall shapes of the extracted glue functions are similar with both methods (see figure 1); in both channels they show a rather narrow low-frequency peak below 500–1000 cm^{-1} and a broad hump up to 3000–4000 cm^{-1}. Despite the similarity of the two channels, a closer inspection shows that the doping dependency is just opposite, with the B_{1g} glue function rapidly decreasing with doping above the optimal, while the glue function extracted from the B_{2g} spectra stays roughly constant. More than this, one can notice that the frequency above which the B_{2g} glue functions vanishes shrinks passing from the 3000–4000 cm^{-1} range, at x = 0.15, to about 2000 cm^{-1}, when x = 0.25. This points to the natural interpretation that spin waves underly the B_{1g} glue function at all frequencies, while charge excitations are responsible for the low/intermediate-frequency scattering in the B_{2g} channel. In this channel spin waves also contribute, but mostly at high frequencies. When doping suppresses the spin-mediated scattering, the overall B_{1g} glue decreases as well as the high-frequency part of the B_{2g}. The marked difference of the doping dependence in the two channels is also visible from both the low-frequency and total spectral weights of the glue functions (see figure 3). A marked increase of the low-energy excitations involved in the B_{2g} channel is also clear from figures 3(c) and (d).

The glue functions extracted here are in overall agreement with those obtained from other techniques. A peaked feature below 1000 cm^{-1} is also present in optical experiment [39, 40] and in low-energy spin excitations revealed by inelastic neutron scattering (INS) [47, 48]. At higher energies (up to 0.3–0.4 eV) broad humps are found in optical spectra like those obtained within our analysis, which can be identified as due to spin excitations both from neutron scattering [49] and from resonant inelastic x-ray scattering (RIXS) [50, 51].

At the same time there is increasing evidence from both NMR [11] and RIXS [26, 27, 29, 30] that CDW are ubiquitous low-energy excitations in cuprates. It is therefore natural to identify them with the excitations involved in the B_{2g} channel.

Our results also display a general agreement with theoretical [41] and experimental [42], ARPES results in Bi_{2}Sr_{2}CaCu_{2}O_{8+x} samples. In particular there is full consistency concerning the coexistence of two distinct collective modes and with the softening of the mode(s) near the ‘hot’ antinodal region. It is noticeable that this agreement is present despite the obvious differences between the Raman response and the electron self-energy. In particular the former is resolved with respect to the electron momentum, while it is integrated over all the momenta of the bosons dressing the single-particle fermionic propagator. On the other hand, the Raman response obeys some (partial) cancellations when the boson momenta and the Raman form factors interplay [23]. From [42] one sees that the mode, which softens in the antinodal region, gets more strongly coupled in the underdoped region and could be identified with our spin-fluctuation mode. The (momentum averaged) energy of the other mode stays more constant over the electronic momentum space and it acquires relatively more weight increasing the doping above optimal.

Figure 4. Main panel: Temperature dependence of the inverse of the effective slope of the glue function at low frequency, 1/\text{eff}, for different channels (B_{1g}, circles; B_{2g}, triangles) and doping levels (x = 0.15, yellow; x = 0.20, pale green; x = 0.25, dark green). Here, s_{eff} has been computed from the M_{g} fitted using the SVD method. Similar results are obtained with the MRD. Inset: the same data in a log–log scale, highlighting a seemingly \text{T}^{3/2} behavior (dashed line).
doping. This leads to the identification of this mode with our mixed phonon-CDW mode, which becomes prominent when doping increases above the optimal level. Concerning the temperature dependence, INS find below \( T_c \) a sharpening of the low-energy part of the spin-fluctuation spectra (see, e.g. [43]), accounting for the clear observation of this mode in the ARPES spectra ([41, 42]) below \( T_c \) (the contribution of the other broad featureless, nearly temperature independent part of the spin fluctuations being naturally buried in the broad incoherent parts of the electron self-energies). The softening of the charge modes extracted (mainly) from the Raman (\( B_{2g} \)) spectra is only expected to occur near the antinodal region and therefore in ARPES may be overshadowed by the softening of the other spin mode. Our selectivity in momentum allows instead to disentangle the contributions of the modes to the different Raman channels and to detect a substantial softening of the charge mode as well.

Our work focuses on temperatures above \( T_c \) and finds a distinct temperature behavior for the overall and the low-energy parts of the spectra. Specifically, we notice a rather weak dependence of the overall spectra, which becomes more pronounced in the low-frequency weight. This indicates that the excitations might have a low-frequency component with a marked temperature dependence. Indeed, as explained in section IV, exploiting analytic low-frequency expansions of the memory function and of the glue functions extracted, e.g. via the SVD method, we are able to identify a substantial temperature dependence of the imaginary part of the memory function at zero frequency. This is related (see equation (6)) to the slope of the Raman response near \( \omega = 0 \). In turn, this is related to the effective slope of the glue function \( s_{\text{eff}} \) defined in equation (7), which is a rather robust quantity extracted from a weighted average of \( \alpha^2 F(z)/z \) that is only weakly sensitive on the oscillatory character of the SVD fitted glue functions.

These strong temperature dependencies at low frequency are suggestive of some form of critical behavior of the mediating excitations. If for a while we borrow the analytic expressions of the glue functions calculated in [23], we find \( m^* g \approx \lambda s_{\text{eff}} \), where \( m \) is the mass of a collective mode (i.e. the minimum energy needed to excite it) and is proportional to the square of the inverse correlation length of the corresponding fluctuations, while \( g \) represents the coupling between fermionic quasiparticles and the collective modes. Thus, although in this work we purposely avoided any assumption on the mediator, we interestingly find that our results support a marked temperature dependence of its mass, as it is seen in figure 4.

Within a quantum critical scenario, the masses are expected to vary linearly with \( T \) in the quantum critical regime, and saturate in the quantum ordered regime. Of course drawing any conclusion on this dependence with three temperatures only is out of question, but a clear indication of nearly critical mediators, with a certain tendency of the mass to flatten as a function of temperature at low \( T \), can still be obtained.

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Appendix A. Multi-rectangle decomposition

In this case the strategy to minimize the functional in equation (5) is based on a histogram decomposition of the glue function, the fitting parameters being the heights of the histograms, in the \( N \) different frequency intervals (bins) in which the frequency axis is partitioned, and the prefactor \( \chi_0 \) (that also include all multiplicative parameters needed to match the theoretically calculated response with the measured one). The fitting procedure searches for a local minimum, beginning from a starting guess, and using the direct search algorithm of Hooke and Jeeves [55–58].

Using as starting point equation (2), we divide the integration range into \( N \) non-overlapping intervals as follows:

\[
M''(\omega) = \sum_{a=1}^{N} \int_{a_i}^{a_{i+1}} dz \, K(\omega, z) \alpha^2 F(z), \quad (A.1)
\]

where \( j = 1, \ldots, N \), and to avoid confusion with the frequencies \( \omega_i \) at which the Raman response is experimentally measured, we called here \( a_0 \) and \( b_0 \) the frequencies identifying the \( \alpha \)-th bin, with \( a_0 < a_{i+1} = b_0 \). In every integration interval of equation (A.1), the glue function is to be considered a constant (the height of the bin) and can be taken out the integral and considered as a multiplicative factor indicated with \( c_a \), so we can rewrite the equation (A.1) in a matrix form

\[
M''(\omega) = \sum_{a=1}^{N} c_a \Delta M''_{a}, \quad (A.2)
\]

with

\[
\Delta M'_{a} \equiv \int_{a_i}^{a_{i+1}} dz \, K(\omega, z) .
\]

The integration domain in equation (2) extends between 0 and \( \infty \). In equation (A.1) we have taken \( b_0 = 8000 \text{ cm}^{-1} = \omega_{\text{max}} \) assuming that the integrand vanishes at higher frequencies. The minimum frequency (acting as a natural cutoff within our procedure) was typically taken as \( a_i = 10 \text{ cm}^{-1} = \omega_{\text{min}} \). Rather than adopting a homogeneous mesh, with constant \( b_0 - a_0 \), we adopted a logarithmic mesh, with constant \( b_0 / a_0 \), to enhance the sensitivity at low frequency. The value of the integral in the bin with extremes \( [a, b] \) appearing in equation (A.1), with \( a, b > 0 \), is

\[
\Delta M''(\omega) = 2T \omega^{-1} \alpha^2 F(\alpha) \left\{ \begin{array}{l} 2\omega \ln \frac{\sinh(\hat{b})}{\sinh(\hat{a})} \\
\Xi(\hat{a} + \hat{\omega}) - \Xi(\hat{a} - \hat{\omega}) - \Xi(\hat{b} + \hat{\omega}) + \Xi(\hat{b} - \hat{\omega}) \\
- \sum_{k=1}^{\infty} \frac{1}{2k^2} \left[ \Lambda(2k\hat{a} + 2k\hat{\omega}) - \Lambda(2k\hat{a} - 2k\hat{\omega}) \right] \\
+ \sum_{k=1}^{\infty} \frac{1}{2k^2} \left[ \Lambda(2k\hat{b} + 2k\hat{\omega}) - \Lambda(2k\hat{b} - 2k\hat{\omega}) \right] \end{array} \right\}
\]
when \( bT \leq 6000 \) functions. The eigenfunctions \( \omega \alpha \) and \( 8000 \omega \alpha \) are ign, treating the divergence in the sense of the principal value, have to be considered. When the results for the integral (A.3) the integrand function is replaced with a continuous function is known at discrete points, to calculate \( P_z M_z \) and \( \omega \alpha \), and the last two lines contain rapidly convergent series. For numerical reasons, we approximated \( \ln(\sinh|x|) \approx |x| - \ln(2) \) when \( |x| > 40 \).

Once the imaginary part of the memory function is obtained, the real part is achieved through the KK transformation

\[
\rho(\omega) = \rho(\omega) \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{M''(\xi)}{\xi - \omega} \, d\xi.
\]  

(A.3)

Since the \( M''(\omega) \) function is known at discrete points, to calculate the integral (A.3) the integrand function is replaced with a continuous broken line obtained by joining the heights of the bin with straight lines. So the analytical form of \( M''(\omega) \) to be included in (A.3) will look like \( M''(\xi) = A\xi + B \), in which the coefficients \( A \) and \( B \) are calculated within each bin. To calculate the integral (A.3) we have subdivided the integration interval in subintervals which are delimited by the same point where (A.3) we have subdivided the integration interval in subintervals (A.3) for the subinterval delimited by, say, \([\omega_1, \omega_2]\] is straightforwardly

\[
A(\omega_2 - \omega_1) + (A\omega + B) \ln \left| \frac{\omega_2 - \omega}{\omega_1 - \omega} \right|,
\]

while in the latter case two neighboring bins \([\omega_1, \omega - \xi]\] and \([\omega + \xi, \omega_2]\] treating the divergence in the sense of the principal value, have to be considered. When the results for the two neighboring bins are summed, the divergent part cancels and the same expression is obtained.

We have to remark that, in general, multiple choices of \( \alpha^2 F(\omega) \) yield the same memory function \( M'' \), due to the presence of nonzero solutions to the matrix equation \( \sum_{\alpha=1}^{N} \Delta M''_{\alpha} c_{\alpha} = 0 \). It is therefore impossible to fit an unique glue function \( \alpha^2 F(\omega) \) without imposing additional constraints (see, e.g. [59]). We restrict the possible glue functions by imposing the following constraints:

1. \( \alpha^2 F(\omega) \geq 0 \), and therefore \( c_{\alpha} \geq 0 \);
2. The initial guess for the glue function in the minimization procedure has vanishing spectral weight for \( \omega > 4000 \) cm\(^{-1} \), assuming that the collective modes live at lower frequencies.

Such constraints do not completely avoid the uncertainty about the glue function. However, the relevant features of the calculated glue function are robust enough, upon varying the doping and the temperature, as discussed in section III.

We performed our calculations comparing the results obtained with \( N = 8, 12, \) and 25 bins, to ensure that the main features of the glue function were robust with respect to variations in the MRD scheme.

**Appendix B. Singular vector decomposition**

In this section we describe in a detailed way the SVD-based fitting procedure. The first step is to discretize the kernel \( K(\omega, z) \) in equation (3). This is done by using two (possibly different) meshes \( \{w_i\} \) and \( \{z_j\} \). Logarithmic meshes, or combinations of linear (at small frequencies) and logarithmic (at large frequencies) meshes, can be used. The integration measure is associated to the kernel, so that equation (2) becomes:

\[
M'' = \sum_j K_{ji} \alpha^2 F_j .
\]

(B.1)

The SVD, although computationally demanding, provides the best low-rank approximation of the kernel \( K \), a result known as the Eckart–Young theorem. In this sense, it is the ideal tool to approximate solutions of integral equations. However, a naïve application of the SVD to the kernel in equation (B.1) produces basis vectors \( \phi_\alpha \) with undesirable properties. (1) On physical grounds, we require that the glue function goes to zero at large frequency (at least as \( 1/z \), see
Figure B2. (a) Fit of \( \chi'' \) using different \( \chi_0 \) values. We show the goodness of fit, as measured by \( \Delta^2 \) (see equation (5)), and the ratio between the area under the glue function for \( \omega \geq 2500 \text{ cm}^{-1} \) to the total glue function weight \( W_{\text{max}} \). Best fit is obtained for \( \chi_0 \sim 17 \). (b) Smaller values of \( \chi_0 \) do not yield a good fit, with \( \chi''_0 \) being systematically smaller than the experimental susceptibility. (c) The fitted glue function increases its total area, in particular at high frequencies, as \( \chi_0 \) is increased. The lines shown in panel (b) (or (c)) correspond to the best fit value \( \chi_0 = 17 \) (blue thick line) and the \( \chi_0 \) values shown in panel (a), smaller (or larger) than 17 (thin lines). All points have been computed using the same mesh (\( N_w = 536, N_z = 507 \)) and cutoff (power-law, see equation (B.3) with \( \Omega = 3000 \text{ cm}^{-1} \) and \( \gamma = 4 \)).

Figure B3. (a) Magnitude of the singular values \( \sigma_\alpha \) of the kernel \( K_0(\omega, z)\psi(\zeta) \), at three different temperatures, normalized by the value of the first SV. (b) Number of singular values \( \sigma_\alpha \) larger than \( 10^{-6} \times \sigma_1 \) as a function of temperature. In both panels the same mesh (\( N_w = 536, N_z = 507 \)) and cutoff (power-law, see equation (B.3) with \( \Omega = 3000 \text{ cm}^{-1} \) and \( \gamma = 4 \)) was used.

Figure C1. The red points are \( M(T, x) \) using the SVD procedure, as a function of temperature and doping, for the two channels. The surfaces represent the fits \( M(T, x) = \sqrt{T} (a + bx) \). The surface parameters are \( a = 190 \) and \( b = -570 \) for the \( B1g \) channel, while they are \( a = 0 \) and \( b = 300 \) for the \( B2g \) channel, where both \( a \) and \( b \) are in \( \text{K}^{1/2}\text{cm}^{-1} \) units.
However, the integration kernel $K$ is long-ranged, in the sense that it is nonzero for large $\omega$ and $z$. As a result, the singular vectors $\phi_\alpha(z)$ do not go to zero for large $z$, making high important to constrain $\alpha^2 F$ to be small at large frequencies. (2) Even worse, the kernel $K(\omega,z)$ diverges as $\sim 1/z$ for $z \to 0$; therefore, the SVD of the kernel produces basis vectors $\phi_\alpha$ which also divergent in $0$. This is in sharp constrast with the fact that the glue function should go to zero fast enough to regularize the integral. In order to ensure the convergence of the integral, equation (2). As described in Appendix A, within the MRD approach these two problems are easily solved by choosing bins with lower edges larger than zero (thus providing a cutoff to the integral) and upper edges smaller than a maximum frequency $\sim 8000 \text{cm}^{-1}$. In the SVD context, the solution to these problems requires to expand a properly regularized kernel instead of the original one; we will detail our procedures in the next sections.

### B.1. Large frequencies

In order to perform numerically the integral in equation (2), some cutoff frequency $\Omega$ has to be introduced. Due to the long-range nature of the kernel, the $\phi$ eigenfunctions are strongly dependent on the cutoff. Therefore, the choice of the cutoff should be motivated by physical reasons, and we have to check that the results are only weakly dependent on the particular cutoff choice.

Let us note that the integration kernel $K(\omega,z)$ goes to zero for $z \gtrsim \omega + \mathcal{O}(e^{-i(\omega-z)})$ (see equation (3)). This means that high frequency ($z > \Omega$) components of the glue function do not contribute to the memory functions at low frequency ($\omega < \Omega$) and, vice versa, that only the low-frequency part of the glue function can be fitted from low-frequency data.

We can explicitly set to zero the high-frequency part of the glue function introducing a cutoff function $q(z)$ as

$$q(z) = \frac{1}{1 + (z/\Omega)^2}, \quad (B.2)$$

where $q(z)$ is 1 at low frequencies and goes to zero at high frequencies, and $f$ does not diverge in the $z \to \infty$ limit. Many choices for the cutoff function $q(z)$ are possible, for instance a power law decay

$$q(z) = \frac{1}{2} \left[ 1 + \tanh \left( \frac{\Omega - z}{\Delta} \right) \right], \quad (B.3)$$

or an exponential decay, e.g.

$$q(z) = \frac{1}{2} \left[ 1 + \tanh \left( \frac{\Omega - z}{\Delta} \right) \right],$$

for suitable parameters $\Omega, \gamma$ and $\Delta$. We find that the results depend only weakly on the cutoff, as the support of the fitted glue functions is concentrated before $2000-3000 \text{cm}^{-1}$. Therefore, we will write $M''(\omega) = \int dz \ K(\omega,z) q(z)f(z)$ and fit $f(z)$ to the experimental data using the cut off kernel $K(\omega,z)q(z)$. After a function $f(z)$ has been determined, the corresponding glue function is obtained from equation (B.2).

### B.2. Small frequencies

The divergence in $z = 0$ of the kernel implies that the right eigenfunctions (which are used as a basis function for the glue function) are divergent in zero. Since $\alpha^2 F(0) = 0$, we want to approximate the glue function using a set of functions for which $\phi_\alpha(0) = 0$. This can be accomplished by expanding a suitable regularized kernel instead of the original one. We can write:

$$K(\omega,z) = K_R(\omega,z) + K_D(\omega,z), \quad (B.4)$$

where $K_R$ is regular in $z \to 0$, and $K_D(z) = 4\pi T z$. We can use the SVD decomposition for the regular part of the kernel, and study the effect of the second term on the solution. Of course, $K_D(z)$ is still divergent, but the right eigenfunctions of $K_R$ go to zero fast enough to regularize the integral.

### B.3. Rewriting the integral equation

Putting together the cutoff definition, equation (B.2), and the decomposition into regular and divergent part, equation (B.4), we are able to recast the initial equation (2) in the following form:

$$M''(\omega) = \int dz \ [K_R(\omega,z)q(z)f(z) + \int dz \ [K_D(\omega,z)q(z)f(z). \quad (B.5)$$

We then perform the SVD on the new kernel $K'(\omega,z) \equiv [K_R(\omega,z)q(z)]$, rather than on the original kernel $K(\omega,z)$, as follows:
\[
K_R(\omega, z)q(z) \approx \sum_{\alpha=1}^{N} \sigma_{\alpha} \psi_{\alpha}(\omega) \phi_{\alpha}(z), \tag{B.6}
\]

where \(N\) is the number of singular vectors we use to approximate the kernel. We can hence rewrite equation (B.5) by expanding \(f(z) = \sum_{\alpha} c_{\alpha} \phi_{\alpha}(z)\), obtaining
\[
M''(z) = \sum_{\alpha} c_{\alpha} A''_{\alpha}(z),
\]

where
\[
A''_{\alpha}(\omega) = \sigma_{\alpha} \psi_{\alpha}(\omega) + \delta\sigma_{\alpha},
\]
\[
\delta\sigma_{\alpha} = \int dz \ K_R(z)q(z)\phi_{\alpha}(z).
\]

The real part of the memory function, \(M'(z)\) can be obtained as a function of the coefficients \(c_{\alpha}\) by taking the KK transform of the functions \(A''_{\alpha}(\omega)\) (or, equivalently, of the \(\psi_{\alpha}(\omega)\) functions). Equation (4) is then obtained by plugging the expressions for \(M'\) and \(M''\) into equation (1).

### B.4. Fitting procedure

A simultaneous fit of both \(\chi_0\) and the expansion coefficients \(\{c_{\alpha}\}\) is often unstable, with \(\chi_0\) being pushed towards very large values. Therefore, we decided to fit the coefficients \(c_{\alpha}\) at fixed \(\chi_0\) and to systematically study the effect of different \(\chi_0\). As explained in the main text, the coefficients \(\{c_{\alpha}\}\) which fix the glue functions are obtained by the minimization of a function \(\Delta^2(\{c_{\alpha}\})\), i.e. the square distance \(\Delta^2\) between the theoretical response function and the experimental one.

It is useful to introduce the primitive of the glue function
\[
W(z) = \int_{z}^{\infty} dz' \alpha^2 F(z'),
\]

and \(W_{\text{max}} = W(z_{\text{max}})\), with \(z_{\text{max}}\) being the largest frequency in the \(z\)-mesh. Therefore, the quantity
\[
1 - W(z)/W(z_{\text{max}})
\]
equals the fraction of the area under \(\alpha^2 F\) at frequencies larger than \(z\).

Figure B2 shows an example (data corresponds to the \(B_{1g}\) channel, \(T = 105\, K\), doping \(x = 0.25\)). As one can see in panels (a) and (b), small values of \(\chi_0\) do not allow for a good fit, while for \(\chi_0 \gtrsim 17\) the \(\Delta^2\) is generally small. However, we see that using a large \(\chi_0\) produces glue functions with a sizable spectral weight at high frequencies, as it is clear from panels (a) and (c). In this case the choice \(\chi_0 = 17\) gives the best combination of fit quality and reduced spectral weight at large frequencies.

We finally address the question of how to choose the number of singular vectors. The contribution of each singular vector \(\phi_{\alpha}\) to the susceptibility is mediated by the singular values \(\sigma_{\alpha}\). The number of singular vectors which have to be kept into account depends on how fast \(\sigma_{\alpha}\) decreases with the index \(\alpha\). As one can see in figure B3, the decay of the singular vectors strongly depends on temperature. At high temperature, the kernel can be approximated by just using a few terms of the SVD. Hence, the number of singular vectors used to approximate the kernel should decrease with \(T\), too. Moreover, we find that the convergence of the fit gets considerably worse if too many singular vectors are used. A reliable choice is using 40 SVs for \(T \sim 50\, K\), 25 SVs for \(T \sim 100\, K\), and 15 SVs for \(T \sim 200\, K\).
Appendix C. Relating low-frequency behavior of Raman responses and glue functions

C.1 Raman response and memory function

Some informations on the glue function can be directly extracted from the low-frequency behavior of the experimental Raman response function $\chi''$. In fact, let us expand both the real and imaginary parts of the memory function near zero as

\[ M'(\omega) = M'_0 + \mathcal{O}(\omega^3), \]
\[ M''(\omega) = M''_0 + M''_2 \omega^2 + \mathcal{O}(\omega^4), \]

where we exploited the opposite parity of the functions $M'$ and $M''$. Substituting into equation (1) we obtain

\[ \frac{\chi''(\omega)}{\chi_0} = \frac{\omega}{M''_0} + \left( \frac{\omega}{M''_0} \right)^3 (M'_1 + 1)^2 - M''_0 M''_2 + \mathcal{O}(\omega^5). \]  

(C.1)

Therefore, the low-frequency behavior of $\chi''$ is controlled by the imaginary part of the memory function, computed at zero frequency. As described in the main text, $M''_0$ is found to have a marked dependence on temperature and doping, as described by equation (8) and shown in figure C1.

The expansion equation (C.1) allows an estimate of the Raman response function $\chi''(\omega)$ in the low-frequency limit. Figure C2 shows how the experimental $\chi''(\omega)$ computed at $\omega = 50$ cm$^{-1}$ (the lowest nonzero frequency available for all the datasets) compares with the first order expansion $\chi''(\omega)/M''_0$, where $M''_0$ can be either the value computed by integrating the fitted glue functions, or the value extracted from the fits in equation (8). The agreement is far from perfect, although the correct trend is respected. We have to remark that the single experimental points used in the comparison can be strongly influenced both by the limited number of data points at low temperature, by random instrumental errors and/or by the presence of phonon excitations. As a consequence, the scaling results, equation (8), obtained using $M''_0$ (arising from global fits to the data) are much more consistent than the ones obtained with $\chi''(\omega)$ alone.

C.2 Memory function and glue function

Equation (2) can be used to relate $M''_0$ and the glue function as follows

\[ M''_0 = \int_0^\infty dz \ K(0,z) \alpha^2 F(z) \]

= \[ 2\pi \int_0^\infty dz \ \frac{z}{2T} \ \text{cosech}^2\left(\frac{z}{2T}\right) \alpha^2 F(z). \]

The kernel $K(0,z)$ has a 1/z divergence at $z = 0$ and dies exponentially as $\exp(-z/T)$. Therefore, the support of $K(0,z)$ is essentially the interval $z \in [0,T]$, so that $M''_0$ is a measure of the spectral weight of the glue function at frequencies $\omega \lesssim T$. In particular, at low temperature, the slope at zero frequency of the glue function is the dominant contribution in the integral. This suggests the definition of the quantity $s_{\text{eff}}$.

\[ s_{\text{eff}} = \frac{3M''_0}{4\pi T^2}. \]  

(C.2)

It is easy to show that, in the $T \rightarrow 0$ limit, $s_{\text{eff}}$ coincides with the slope $\text{d}\alpha^2 F(\omega)/\text{d}z$ at $z = 0$. This can be seen by expanding in series the glue function around $z = 0$ as $\alpha^2 F(z) = c_1 z + c_2 z^2 + c_3 z^3 + \ldots$. The coefficients $c_i$ depend in principle on both temperature, doping and channel. Then, we can compute the contribution of each term to $M''_0$ (this may not be a convergent series, it depends on how fast the coefficients $c_i$ decay with $n$; however, we assume that it is at least an asymptotic series for low enough $T$):

\[ M''_0 \sim \sum_{n \geq 1} \int_{0}^{\infty} K(0,z) c_n^2 \text{d}z \]

= \[ \sum_{n \geq 1} 2\pi (2T)^{n+1} \int_{0}^{\infty} \text{cosech}^2(y) y^{n+1} \text{d}y. \]

The function $J(y) = \int_{0}^{\infty} \text{cosech}(t) t^n \text{d}t$ is well defined for complex-valued arguments in the domain $\text{Re}(x) > 1$, and can be expressed as $J(y) = \text{Re}(x) \Gamma(n+1)$, $\text{Re}(x) > 1$, where $\Gamma(x)$ is Euler’s gamma and Riemann’s zeta functions, respectively. The first few values of the integral are $J(2) = \pi^2/6$, $J(3) = 3\zeta(3)/2$ and $J(4) = \pi^4/30$. This leads to the result

\[ M''_0 \sim \sum_{n \geq 1} \left( 2\pi (2T)^{n+1} \right) (n+1) \]

= \[ 4\pi T \sum_{n \geq 1} c_n T^n \times \zeta(n+1) \Gamma(n+2) \]

= \[ 4\pi^3 \frac{T^2 c_1}{3} + 24\zeta(3) T^3 c_2 + 16\pi^5 \frac{15}{15} T^4 c_3 + \ldots \]  

(C.3)

Therefore, $s_{\text{eff}} \sim c_1$ if the first term dominates the series in equation (C.3). Using the known value of the integral $J(2)$, we see that another way to define $s_{\text{eff}}$, equivalent to equation (C.2), is

\[ s_{\text{eff}} = \int_{0}^{\infty} \text{d}z \left[ \frac{\alpha^2 F(z)}{z} \frac{1}{\text{cosech}^2\left(\frac{z}{2T}\right)} \right]. \]

This expression represents an average of the quantity $\alpha^2 F(z)/z$ with a fast-decaying weight $\approx (z/T)^2 \exp(-z/T)$ for $z \gg T$. We show in figure C3 how the values $M''_0$ computed from the SVD-fitted glue functions can be used to estimate the low frequency behavior of the Raman response function $\chi''(\omega)$ (panels (a) and (c)), and the effective slope $s_{\text{eff}}$. We see that the extrapolated slope of $\chi''$ fits quite well with the experimental data, even when the data points are scarce. The effective slope of the glue function, instead, is systematically smaller than the slope of the fitted glue functions at $z = 0$. This is due to the glue functions being concave. However, $s_{\text{eff}}$ appears to be roughly proportional to $\alpha^2 F(z)/z$ on doping. In particular, this suggests that the dependence of $s_{\text{eff}}$ and $\alpha^2 F(z)/z$ on doping are very similar. Figure 4 in the main text reports the temperature dependence of the inverse effective slope $s_{\text{eff}}$, highlighting the strong temperature dependence of the low-frequency glue function, possibly related to the quantum critical behavior of
the interaction mediator [23]. Although the plot is limited to three temperatures, this quantity seems to be quite compatible with a power-law dependence $s_{\text{eff}} \sim T^{1 \pm 2}$, as it is seen from the inset of figure 4, calling for further investigation.

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