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# The role of magnetism in forming the $c$ -axis spectral peak at $400\text{ cm}^{-1}$ in high temperature superconductors

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## Abstract

We discuss the peak at  $400\text{ cm}^{-1}$ , which is seen in  $c$ -axis conductivity spectra of underdoped high temperature superconductors. The model of van der Marel and Munzar, where the peak is the result of a transverse plasmon arising from a low frequency conductivity mode between the closely spaced planes, fits our data well. Within the model we find that the temperature dependence of the peak amplitude is controlled by in-plane scattering processes. The temperature range where the mode can be seen coincides with  $T_s$ , the spin gap temperature, which is lower than  $T^*$ , the pseudogap temperature. As a function of temperature, the amplitude of the mode tracks the amplitude of the 41 meV neutron resonance and the spin lattice relaxation time, suggesting to us that the mode is controlled by magnetic processes and not by superconducting fluctuations which have temperature scale much closer to  $T_c$ , the superconducting transition temperature.

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## 1. Introduction

The first infrared measurements on ceramic samples of high temperature superconductors early in 1987 showed two unexpected features. In addition to the well known optical phonons of the perovskite lattice, two new features could be seen. The first was a dramatic reflectance edge that appeared below the superconducting transition temperature and the second, seen in many samples with more than one copper–oxygen plane, was a peak in the  $400\text{ cm}^{-1}$  region, much broader than a typical phonon. The peak frequency of the feature increases with doping but we will refer to it as the  $400\text{ cm}^{-1}$  peak which is the position in underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{6.60}$ . The reflectance edge was soon explained as a plasma edge, resulting from the zero crossing of the real part of the dielectric function where the positive

contribution of the ionic displacement current cancels the negative current due to the superconducting condensate [1]. Since the super-current is a current of pairs, the plasma edge has been termed the Josephson plasma edge [2,3]. However, the appearance of the plasma edge is a classical effect and it would be present in any conductor where the scattering rate is sufficiently small,  $1/\tau \ll \omega_p$ .

The peak at  $400\text{ cm}^{-1}$  was ignored in the literature until the work of Homes et al. [4–7] where its properties were described in some detail. Recently, the two phenomena have been connected by suggestions that the  $400\text{ cm}^{-1}$  peak is a *transverse* plasma resonance associated with a coupling between the closely spaced planes of the bilayers by van der Marel and by Munzar et al. [8–10]. A transverse plasma resonance occurs in situations where two or more plasmas are geometrically separated. This is in contrast to a multi-component plasma, for example one of light and heavy holes in a semiconductor, which gives rise to a single longitudinal mode with a frequency that depends on the two masses. On the other hand, if the light and heavy hole

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plasmas phase-separate into droplets of a heavy hole plasma within a light hole medium two longitudinal and one transverse plasma resonance will be seen [11,12].

Shibata and Yamada have reported the presence of two longitudinal Josephson plasma resonances in  $\text{SmLa}_{1-x}\text{Sr}_x\text{CuO}_{4-\delta}$  through the observation of sphere resonances [13]. Powdered crystals, suspended in an optically transparent matrix, show a resonance absorption at  $\omega = \omega_p\sqrt{3}$  where  $\omega_p$  is the Josephson plasma frequency [13,14].

The material Shibata and Yamada used to obtain the two separated plasmas has single copper–oxygen planes separated by alternating  $\text{Sm}_2\text{O}_3$  and  $(\text{La}, \text{Sr})_2\text{O}_{2-\delta}$  blocking layers. When the material becomes superconducting two resonances are seen. Both appear at  $T_c$  and follow mean-field temperature dependencies although the two curves are not identical as shown in the inset of Fig. 2 of Ref. [13]. The authors attribute the resonances to the two different series Josephson junctions in the two different blocking layers. The transverse plasma resonance in this material has also been seen [15–17].

Kojima et al. found that a 7 T magnetic field, applied along the  $c$ -axis, has the effect of introducing a new mode at low frequency which is interpreted in terms of transverse plasma resonance modulated by vortices [18]. The presence or absence of a vortex between the layers gives rise to two kinds of Josephson junctions which in turn leads to a transverse Josephson resonance as originally proposed by van der Marel and Tsvetkov [19]. Kojima et al. also found that the spectral weight of the  $400\text{ cm}^{-1}$  mode is enhanced in a magnetic field by some 20% while at the same time the spectral weight associated with the superconducting condensate is reduced. They suggest this is due to a transfer of spectral weight from the low frequency modes to the  $400\text{ cm}^{-1}$ , which they assume to be a transverse Josephson mode [18].

Fig. 1 shows the reflectance and the  $c$ -axis optical conductivity of a  $\text{YBa}_2\text{Cu}_3\text{O}_{6.60}$  crystal with  $T_c = 58\text{ K}$  at a series of temperatures in the region of the spectral peak at  $400\text{ cm}^{-1}$  from the work of Homes et al. [5] The rapid buildup of the spectral peak can be seen clearly along with the reduction in the strength of the phonon at  $320\text{ cm}^{-1}$  which corresponds to the out-of-plane vibrations of the planar oxygens. The strength of the chain oxygen peak at  $280\text{ cm}^{-1}$  is only slightly affected by the buildup of the spectral peak. The  $400\text{ cm}^{-1}$  peak can also be seen in the spectra of  $\text{YBa}_2\text{Cu}_4\text{O}_8$ , [21,20]  $\text{Pb}_2\text{Sr}_2(\text{Y}, \text{Ca})\text{Cu}_3\text{O}_8$ , [22] the three layer  $\text{Ti}_2\text{Ba}_2\text{CaCu}_3\text{O}_{10}$ , [23] and the ladder compound  $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$  [24]. A weak feature has been reported in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ , [25,26] and in  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  as well [27]. It is not seen in the one-layer  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .

An important contribution to the debate about the strange phonon anomalies and the peak at  $400\text{ cm}^{-1}$  was made by van der Marel and Tsvetkov [19] and by Munzar et al. [10] This idea had two parts. First, they assumed that the peak was due to a transverse plasmon (intra-bilayer plasmon)

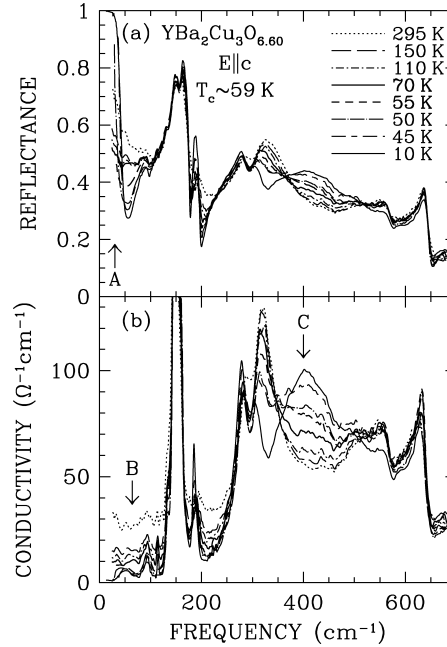


Fig. 1. The reflectance (a) and the optical conductivity (b) at various temperatures of an underdoped sample of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.60}$  for light polarized along the  $c$  axis. The heavy solid line is just above the superconducting transition temperature, the dashed line just below it. Note the appearance of the peak well above the superconducting transition temperature which is  $58\text{ K}$  for this sample. As the peak at  $400\text{ cm}^{-1}$  grows the phonon at  $320\text{ cm}^{-1}$  weakens and shifts to lower frequencies. We interpret this in terms of changing local fields between the planes. The lower frequency phonon at  $280\text{ cm}^{-1}$  is not affected by these fields. The peak at  $400\text{ cm}^{-1}$  cannot be seen above a temperature of  $150\text{ K}$ . The arrow A is the frequency used to estimate the condensate strength, the arrow B the pseudogap depth and the arrow C the strength of the  $400\text{ cm}^{-1}$  peak.

resulting from the coupling between the two planes of the bilayer system. The coupling across the blocking layers is weaker and as a result of two unequal junctions in series, a charge imbalance was set up in the unit cell which alters the local electric fields acting on the ions. Model calculations showed that both the peak at  $400\text{ cm}^{-1}$  and the strengths of nearby phonon lines were reproduced [10]. In particular, the dramatic loss of intensity of the bond-bending mode of the planar oxygens at  $320\text{ cm}^{-1}$  was well accounted for.

The total spectral weight in the  $250\text{--}700\text{ cm}^{-1}$  region is approximately constant as the spectral peak develops. This means that it gains its spectral weight from either the phonons in this spectral region or from a redistribution of the electronic background. In Homes et al. it was assumed that the phonons were the source whereas Bernhard et al. argued, based on a different interpretation of the electronic background, that only 40% of the spectral weight is accounted for by phonons and the remainder from an assumed lower electronic background [28]. As the raw reflectance and ellipsometric data used in Bernhard et al. are nearly

identical, the different conclusions result from different assumptions as to the baseline in the  $400\text{ cm}^{-1}$  region. These anomalies in the phonon spectral weight are strictly a low temperature phenomenon. It was shown by Timusk et al. that at room temperature the strengths of the  $c$ -axis polarized phonons had their expected magnitudes as determined by formal charges on the ions and a shell model based on inelastic neutron spectroscopy [29].

The temperature dependence of the  $400\text{ cm}^{-1}$  mode has been discussed in the literature by several authors. It was pointed out by Homes et al. that while the mode is present above  $T_c$  it appears at a temperature of 150 K, which is much lower than that of the pseudogap and which can be seen at room temperature [6]. Schützmann et al. suggested that the phonon anomalies might be associated with the spin gap in the nuclear spin relaxation rate  $1/(T_1T)$  that occurs at 150 K [7]. Following up on this idea, Hauff et al. showed that Zn substitution had a dramatic effect of destroying the  $400\text{ cm}^{-1}$  peak while at the same time not changing the pseudogap, which they defined as a low frequency depression of conductivity. More recently, it has been suggested that the current  $J_{bl}$  within the bilayers that is responsible for the mode at  $400\text{ cm}^{-1}$  is a Josephson current [10,28]. It is difficult to see how such a current can exist in the normal state up to 150 K. Note however, that the existence of the peak at  $400\text{ cm}^{-1}$  does not require that the intra-plane current  $J_{bl}$  is superconducting as pointed out by Grüninger et al. [9]. We will return to the problem of the interlayer current below.

The focus of the remainder of the paper is on the temperature dependence of the  $400\text{ cm}^{-1}$  mode. First we emphasize that the mode appears at 150 K, well above the superconducting transition temperature. This fact is difficult to reconcile with the idea of a transverse Josephson plasmon unless some kind of 2D superconductivity exists above  $T_c$  in the planes. We discuss the evidence for this kind of superconductivity. Then we examine models within the van der Marel/Munzar (vdMM) picture where the oscillator that represents the intra-bilayer current  $J_{bl}$  is allowed to have a width. Next, we focus on the temperature dependence of the mode. We note, as first suggested by Schützmann et al. that the mode follows the temperature dependence the NMR  $1/(T_1T)$  connecting the mode intimately to the spin gap and magnetism but we find that the mode intensity follows even more closely the intensity of the 41 meV neutron mode. In contrast, the Knight shift temperature scale is higher and more closely related to the pseudogap in the conductivity. Finally, we offer some speculations on what might cause this intimate connection between intra-bilayer transport and magnetism.

## 2. Results and discussion

The idea that the transverse plasmon giving rise to the

$400\text{ cm}^{-1}$  peak is the result of the Josephson effect connecting the two superconducting planes has some attractive features. The model of Munzar et al. shows that there is a narrow peak near zero frequency corresponding to a current  $J_{bl}$  between the closely spaced layers. It first appears at 150 K and grows rapidly at  $T_c$ . However, this requires the existence of some kind of superconductivity in the planes at temperatures as high as 150 K. Moreover this superconductivity has to be nearly fully developed at  $T_c$  with a plasma frequency of  $1000\text{ cm}^{-1}$  at  $T_c$  rising to  $1200\text{ cm}^{-1}$  at 4 K [10]. An obvious source for this above- $T_c$  superconductivity is superconducting fluctuations. However, there is little evidence for strong superconducting fluctuations in the  $ab$ -plane properties over a broad frequency range [30]. The only work we are aware of is the paper by Corson et al., who report a weak effect in underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  with an onset temperature around 92 K (in a sample with  $T_c = 74\text{ K}$ ) and a  $\sigma_2$  magnitude of some 18% of the low temperature value measured in the terahertz region [31]. A similar narrow range of temperatures has been reported by Sugimoto et al. [32] in STM measurements of magnetic flux and by Zhang et al. [33] with measurements of the Nernst effect. While there is evidence for vortices above  $T_c$  we are not aware of any transport measurements reporting  $ab$ -plane superconductivity on the temperature scale of  $T_c$ . Finally the presence of the peak in the non-superconducting ladder compound  $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$  [24] speaks against a direct superconducting origin of the peak. Thus it seems unlikely that superconductivity is the source of the  $400\text{ cm}^{-1}$  peak.

One of the advantages of the model of van der Marel and Munzar [10] is that it allows a study of the transport between the layers in a double-layer system, independent of the properties of the blocking layers. We will use the model to study that transport by investigating the parameters that control the properties of the transverse plasmon. In the original paper it was assumed that the current between the layers was a Josephson current between two coupled superconductors. The only parameter controlling the motion of charge between the closely spaced planes in that picture was the Josephson plasma frequency  $\omega_{bl}$ . Classically, the current was due to a Drude term with zero damping parameter and a plasma frequency  $\omega_{bl}$ .

We will next examine the dynamic properties of the transverse plasmon that gives rise to the  $400\text{ cm}^{-1}$  peak within the Munzar model. In the original model the transverse plasmon was the result of a mode at the origin of zero width and a plasma frequency of  $1200\text{ cm}^{-1}$ . Using the parameters of Ref. [10] we have recalculated the conductivity in the  $200$  to  $700\text{ cm}^{-1}$  region adding larger and larger amounts of width to the mode. Fig. 2 shows the results of this calculation. Comparing this with Fig. 1 we see that broadening the mode in the model matches the increasing temperature in the experimental spectra. In other words the temperature dependence of the observed transverse plasmon can be modeled by a low frequency

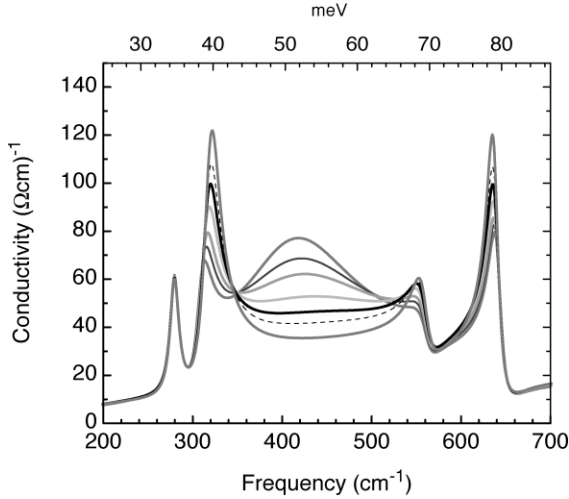


Fig. 2. Model calculation of the influence of mode width on the peak at  $400 \text{ cm}^{-1}$ . Starting from the parameters of Munzar et al. [Ref. [10], top curve, we have added a damping to the mode at the origin. The values for the damping are 25, 50, 100, 150, 200, and  $300 \text{ cm}^{-1}$ . The mode weakens as the damping increases.

Drude-like mode whose width increases approximately linearly from zero to  $300 \text{ cm}^{-1}$  and from low temperature to 150 K. We have also examined the effect on the mode of a shift away from zero frequency. Here the changes are much larger and the center frequency of the mode has to be less than  $50 \text{ cm}^{-1}$  to agree with the experimental data.

As a reference we can look at the scattering rate associated with the in-plane conductivity [34]. With an in-plane plasma frequency of  $7000 \text{ cm}^{-1}$  [34] we can estimate the scattering rates to vary from  $25 \text{ cm}^{-1}$  at 62 K to  $208 \text{ cm}^{-1}$  at 150 K. Comparing these figures with the model shown in Fig. 2 we can predict the size of the  $400 \text{ cm}^{-1}$  peak from the model and the scattering rates calculated from dc transport. These predicted values are shown as crosses in Fig. 3. While the agreement with the observed peak size is rough, at least the calculation shows that in-plane scattering is a possible source of the destruction of the  $400 \text{ cm}^{-1}$  peak as the temperature increases.

We want to follow up the suggestion of Schützmann et al. and investigate the role of magnetism in promoting the intralayer currents. We start by plotting in Fig. 3 several quantities that can be extracted from the  $c$ -axis optical properties. The most important of these for the purposes of the present paper is the amplitude of the  $400 \text{ cm}^{-1}$  mode. We have plotted, as a function of temperature, the conductivity at the maximum of the  $400 \text{ cm}^{-1}$  peak, minus its room temperature value, normalized to its low temperature value based on the 1993 reflectance measurements of Homes et al. [4] It can be seen that the peak first appears at 150 K, some  $90^\circ$  above the superconducting transition, has its most rapid development near  $T_c$  where its amplitude is almost exactly 50% of the saturation value, which it reaches quickly below  $T_c$ .

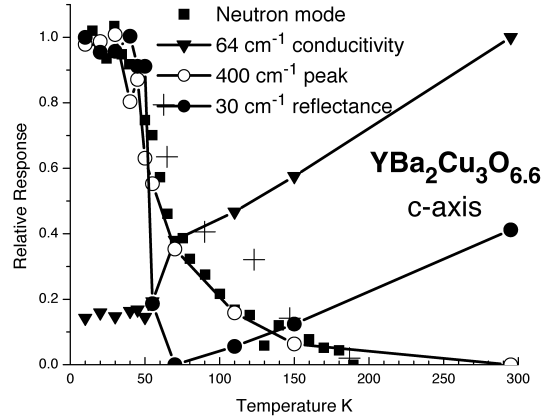


Fig. 3. Response of various quantities as a function of temperature. Three temperature scales can be identified. The largest is around 300 K and is associated with the pseudogap which is approximated as the conductivity at  $64 \text{ cm}^{-1}$ . The next governs the intensity of the  $400 \text{ cm}^{-1}$  peak and neutron mode. It has a value of approximately 150 K. The lowest one is associated with superconductivity and has a value of 60 K in this underdoped sample. We use the  $30 \text{ cm}^{-1}$  reflectance to approximate the superconducting condensate density. The crosses show an estimate of the  $400 \text{ cm}^{-1}$  peak amplitude based on broadening by in-plane scattering.

We have also plotted in Fig. 3 a rough estimate of the superconducting condensate density based on the reflectance at low frequency ( $30 \text{ cm}^{-1}$ ) normalized to unity at 10 K and to zero at minimum reflectance at  $T_c$ . We note that the condensate density rises rapidly at  $T_c$  with a very narrow pre-transition region of no more than 10 K. The slow rise of the low frequency reflectance above  $T_c$  is due to changes in the optical properties associated with the pseudogap.

Also plotted in Fig. 3 is the pseudogap amplitude defined as the conductivity at  $64 \text{ cm}^{-1}$  normalized to unity at room temperature. The pseudogap amplitude drops smoothly from room temperature with a rapid step in the transition region to superconductivity. This step may be due to the formation of the superconducting gap as spectral weight is removed from the low frequency region to the delta function peak at the origin.

We see from Fig. 3 that three temperature scales are involved in the electrodynamics of  $c$ -axis transport. The highest of these describes the pseudogap and has been traditionally called  $T^*$ . At this doping level  $T^*$  is approximately 300 K. The scale associated with the  $400 \text{ cm}^{-1}$  peak is lower and we will call it  $T_s$  for reasons that will become clear below. The phenomena that follow the  $T_s$  scale first appear at 150 K, rise rather slowly as the temperature is lowered, reach about 50% of their final value at the superconducting transition and then rise more rapidly. Finally we have  $T_c$ , the temperature of the transition to a true 3D coherent superconducting state.

To illustrate another phenomenon that seems to follow the  $T_s$  scale we have plotted in the same diagram the amplitude of the 41 meV neutron resonance peak [35]. This

peak is loosely called 41 meV resonance but it occurs at 37 meV in underdoped materials. It was originally thought to be associated with superconductivity since in the optimally doped materials it appeared only below  $T_c$ , but as Fig. 3 shows its temperature scale coincides with the spin gap scale  $T_s$  with an onset at 150 K.

To further explore the various magnetic temperature scales involved in underdoped cuprates we have replotted in Fig. 4 the pseudogap and the  $400\text{ cm}^{-1}$  mode along with two other magnetic quantities, the  $^{63}\text{Cu}$  NMR relaxation rate  $1/T_1T$  and the  $^{63}\text{Cu}$  Knight shift [36]. As pointed out by Homes et al., the depth of the pseudogap follows quite closely the temperature evolution of the NMR Knight shift. This quantity is plotted from the work of Takigawa et al. [36] in Fig. 4. On the other hand we see, as first noted by Hauff et al., the  $400\text{ cm}^{-1}$  peak tracks the evolution of the  $^{63}\text{Cu}$  relaxation rate.

The idea of two pseudogap temperature scales has been used in the NMR literature [37] where the lower scale has been designated  $T^*$  and the higher one  $T_0$ . Here we will not follow that practice but will remain with the more common usage and call the higher scale pseudogap or  $T^*$  scale and will rename the lower scale  $T_s$  or the spin gap scale. From Fig. 4 it seems clear that the peak at  $400\text{ cm}^{-1}$  follows the  $T_s$  scale.

While it appears from Figs. 3 and 4 that the  $400\text{ cm}^{-1}$  mode follows the spin gap there are subtle factors that should be noted. First the temperature dependence of the 41 meV mode matches the  $400\text{ cm}^{-1}$  peak better than the spin relaxation  $1/T_1T$ . In particular, at  $T_c$  there is a rapid rise

in both the 41 meV mode and the  $400\text{ cm}^{-1}$  peak, whereas the spin relaxation rate has no such singularity at  $T_c$ .

Further support for the two temperature scales is obtained from Zn doping studies. It is known from the NMR work of Zheng et al. that 1% Zn doping of  $\text{YBa}_2\text{Cu}_4\text{O}_8$  completely suppresses  $T_s$  while leaving the Knight shift and  $T^*$  unaffected [39,40]. In accord with this, Hauff et al. found that 1.3% Zn substitution of a  $\text{YBa}_2\text{Cu}_3\text{O}_{6.56}$  crystal eliminates the  $400\text{ cm}^{-1}$  peak (and also restores the  $320\text{ cm}^{-1}$  phonon) while not affecting the amplitude of the pseudogap. Thus Zn appears to destroy the spin gap and not affect the pseudogap.

The effect of Zn on the neutron mode is more complicated. From what we have learned from NMR and  $c$ -axis transport we would expect Zn to destroy the neutron mode. However, at least in optimally doped materials, Zn drastically broadens the neutron mode but leaves its spectral weight unchanged up to a concentration of 1.0% [41]. This is in accord with measurements of  $ab$ -plane transport. In  $\text{YBa}_2\text{Cu}_4\text{O}_8$  even 0.425% doping has the effect of dramatically increasing the low temperature scattering rate at  $400\text{ cm}^{-1}$  up to the value that it has in the normal state ( $1200\text{ cm}^{-1}$ ) [38,45]. Thus, it appears that the primary effect of Zn in the spin gap state is to reduce the carrier life time.

We have found from our analysis of the temperature dependence of the  $400\text{ cm}^{-1}$  peak and the vdMM model for the intra-layer plasmon that the  $c$ -axis data can be understood if there is a current  $J_{bl}$  between the closely spaced layers that starts at 150 K and grows rapidly at  $T_c$ . What is the nature of this current? We can rule out several candidate currents because they would require an  $ab$ -plane counterpart. These would include superconducting fluctuations and collective modes that have to do with sliding density waves. The  $ab$ -plane transport shows no diverging conductivity channels in the 150–60 K region in underdoped materials.

The  $c$ -axis dc conductivity matches with the infrared conductivity and the ‘semiconducting’ temperature dependence, at least in the 6.60 doping region, is due to the reduced conductivity as a result of the pseudogap [5]. However, within the vdMM model, this overall poor conductivity does not rule out a highly conducting element in series with a resistive one. From the good fits of the model it is clear that there is a high conductivity element between the closely spaced bilayers and the interbilayer part is poorly conducting.

One possible current path between the bilayers that would be enhanced by long lived magnetic spin flip excitations is shown in Fig. 5. If we assume that the bilayers are antiferromagnetically correlated then a spin flip is necessary to move a hole from layer A to layer B. This means that there is an intermediate state of high energy that has to exist before the antiferromagnetic order is restored. The magnetic barrier splits the two states into an even and odd component and a transition between the two is optically active. Thus we expect a low lying transition, polarized in the  $c$  direction that becomes distinct when the barrier

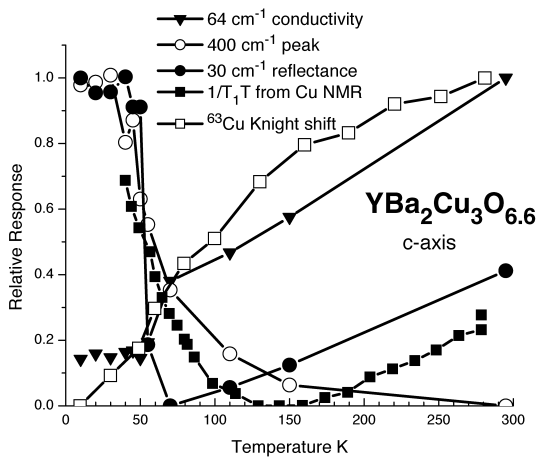


Fig. 4. Two temperature scales seen in NMR. The higher scale seen in the Knight shift plotted as open squares and the pseudogap plotted as triangles and approximated here as the  $64\text{ cm}^{-1}$  conductivity. The Knight shift depression starts at room temperature in this sample with an oxygen content of 6.60. The relaxation time, shown as solid squares, is plotted as  $[1 - 1/(T_1T)]$ . It starts to deviate from the uniform high temperature form at around 150 K and fits better to the  $400\text{ cm}^{-1}$  peak intensity than the Knight shift.



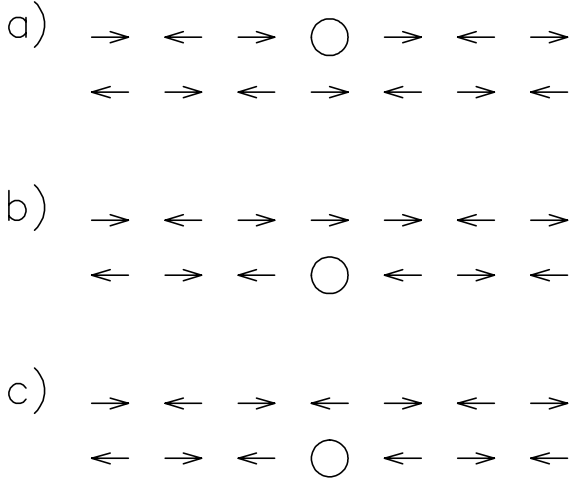


Fig. 5. To transfer a hole from layer (a) to its neighbor in antiferromagnetically ordered planes requires an intermediate state of higher energy (b). A spin flip can restore the ground state (c). We suggest that the system resonates between states (a) and (c) forming an odd and even component. Transitions between these are optically active and may be responsible for the low lying mode giving rise to the intrabilayer current  $J_{bl}$ .

becomes well defined, i.e. when the various states involved become coherent. Another current path is a diagonal second neighbor, one that does not require a spin flip.

The current path shown in Fig. 5 is one of several depending on one's favorite model for the order that develops in the pseudogap or the spin gap states. We are confident that many of these will also contain the ingredients for a mechanism for a current between the closely spaced layers that yields a  $c$ -axis current with properties that do not contradict the experiments.

One problem with any model that involves the 41 meV mode directly is the influence of a static magnetic field. Neutron scattering carried out in magnetic fields shows a reduction of the intensity of the resonance. Dai et al. find that a field of 7 T applied in the  $c$  direction has the effect of reducing the resonance intensity by 20%, while Kojima et al. find, in contrast, that the 400  $\text{cm}^{-1}$  peak grows by some 20% in a comparable field. The field *reduces* the strength of the neutron resonance while it *enhances* the 400  $\text{cm}^{-1}$  peak. Therefore, while the two show the properties of the state that spans a broad region from some 80 K above the superconducting transition down to 0 K, the neutron resonance cannot be a direct cause of the 400  $\text{cm}^{-1}$  peak.

### 3. Conclusion

We have shown here that at least three temperature scales are needed to describe the properties of underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . The largest of these, traditionally termed  $T^*$ , describes the gap that develops in  $c$ -axis transport at low

frequency. This scale also fits the NMR Knight shift as a function of temperature [4,36] and the gaps seen in other spectroscopies such as tunneling and ARPES [37]. It is a broad scale with an onset near room temperature. ARPES has shown that this gap has d-wave symmetry and its appearance in  $c$ -axis transport is a direct result of the matrix elements of the  $d_{x^2-y^2}$  wave functions of the doped holes [42–44].

The second, lower temperature scale, is associated with spin fluctuations—nuclear spin relaxation, and the neutron resonance and the 400  $\text{cm}^{-1}$  peak. We call this scale  $T_s$ . The phenomena associated with this scale become observable tens of degrees above the superconducting transition and have their maximum rate of increase at  $T_c$ . It is a magnetic scale describing  $(\pi, \pi)$  spin fluctuations. It is not clear to us why the intraplane current should develop a narrow Drude-like response when the spin gap appears but we have suggested one possibility that involves the 41 meV neutron mode. Others are possible in the presence of a spin density wave, charge order or dynamic stripes. It is important to note that this magnetic scale  $T_s$  is intimately connected to superconductivity, the phenomena that follow this scale increase rapidly in amplitude at  $T_c$ . Particularly in optimally doped materials it is not possible to separate the scales  $T_c$  and  $T_s$ .

Finally, the properties described by the lowest scale appear abruptly at  $T_c$ , the superconducting transition temperature. Long range superconductivity as shown in our  $c$ -axis experiments by the buildup of spectral weight and by the delta function contribution to the conductivity at the origin follows this scale. There is possibly a fourth temperature scale very close to  $T_c$  where 2D superconducting fluctuations exist.

We find that we can fit the 400  $\text{cm}^{-1}$  peak in the spectra of Homes et al., in agreement with previous work of Munzar et al. [10] to a transverse plasmon arising from current fluctuations between the two planes of the copper-oxygen bilayer. The nature of these fluctuations is not clear to us but they are closely associated with the spin gap. The original suggestion that they are due to 2D Josephson fluctuation in the planes cannot be reconciled with the absence of such fluctuations in  $ab$ -plane transport. Also the presence of the peak in the non-superconducting ladder compound  $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}$  is hard to explain within the Josephson picture.

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