Temperature dependence of the dynamic susceptibility $\chi''(\omega)$ in superconducting $YBa_2Cu_3O_{6.6}$ ($T_c = 53$ K)

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Temperature-dependent neutron-scattering measurements of the dynamic susceptibility $\chi''(\omega)$ in single-crystal $YBa_2Cu_3O_{6.6}$ ($T_c = 53$ K) are reported. At high temperatures ($T \gtrsim 100$ K) a universal dependence of $\chi''(\omega)$ on the ratio ω/T has been observed. A spin pseudogap $\omega_g \cong 10$ meV results in a breakdown in this behavior at temperatures well above T_c. Below ~ 40 K the scattering intensities at energy transfers of $\Delta E = 5$ and 8 meV demonstrate a surprising recovery with decreasing temperature. The behavior of $\chi''(\omega)$ over the range of energies (3–16 meV) and temperatures (1.5–300 K) studied presents a strong constraint on theoretical descriptions of $YBa_2Cu_3O_{6+x}$.

I. INTRODUCTION

Antiferromagnetic spin fluctuations play an important role in many theoretical models of the superconducting and normal-state properties of the high-T_c cuprates. Varma *et al.*[1] suggested that a simple dependence of the imaginary part of the dynamic susceptibility χ'' on the ratio ω/T could be related to the unusual normal-state properties observed in these materials. Subsequent neutron-scattering studies of χ'' in lightly doped, nonsuperconducting $La_{2-x}(Ba,Sr)_xCuO_4$ have found that the integral of χ'' around the antiferromagnetic zone center exhibits a simple scaling dependence on ω/T.[2,3] However, the applicability of ω/T scaling to the description of magnetic dynamics in superconducting samples remains an open issue.

Neutron-scattering studies of magnetic fluctuations in $YBa_2Cu_3O_{6+x}$ have demonstrated the existence of strong dynamic correlations, broadly peaked about the antiferromagnetic zone center, at all oxygen concentrations up to and including optimal doping ($x \sim 1$).[4–7] In superconducting samples a spin pseudogap, which appears to be strongly dependent on oxygenation, is observed at low temperatures.[8–10] The possibility of a simple relationship between χ'' and ω/T that incorporates the effects of this pseudogap has encouraged more detailed studies of the temperature dependence of the magnetic scattering in this system.

In this paper we report temperature-dependent measurements of the dynamic susceptibility χ'' in single-crystal $YBa_2Cu_3O_{6.6}$. Measurements were taken at temperatures ranging from 1.5 to 300 K at energies between 3 and 16 meV. In contrast to the energy dependent normalization employed in recent ω/T scaling studies of $La_{2-x}Sr_xCuO_4$ (Ref. 2) and $YBa_2Cu_3O_{6+x}$ (Ref. 11), we have observed a universal dependence of χ'' on ω/T (no energy dependent prefactor) at temperatures above $T \sim 100$ K. Below ~ 100 K, a spin pseudogap results in a suppression of $\chi''(\omega)$ for energies $\omega < \omega_g \sim 10$ meV. Our most surprising result is the observation, at temperatures below 40 K, of a recovery in the scattering intensity with decreasing temperature at energy transfers of 5 and 8 meV.

II. EXPERIMENTAL DETAILS

The crystal used for these measurements, labeled No. 30b, was recently the subject of a detailed neutron-scattering study by Tranquada *et al.*[12] It was grown at the Institute for Molecular Science,[13] and has a $\sim 1.6°$ mosaic and a volume of ~ 1 cm^3. ac susceptibility measurements indicate a superconducting transition temperature of $T_c = 53$ K. Lattice constants measured on a calibrated spectrometer at room temperature are $a = 3.842$ Å, $b = 3.878$ Å, and $c = 11.738$ Å.

The measurements were performed on the H7 triple-axis spectrometer at Brookhaven National Laboratory's high-flux beam reactor. The (002) reflection of pyrolytic graphite was used to monochromate and analyze the incident and scattered beams. The data were taken using a 40′-40′-S-80′-80′ collimator configuration. Except where noted, measurements were done using a fixed final energy of 14.7 meV. A pyrolytic graphite filter was used to remove higher-order (λ/n) contamination from the scattered beam. The crystal was mounted on the cold finger of a Displex He refrigerator in an aluminum can backfilled with helium exchange gas. The sample was oriented with the $[1\bar{1}0]$ axis normal to the scattering plane to allow the study of momentum transfers $\mathbf{Q} \in (h,h,l)$.

The magnetic fluctuations in metallic $YBa_2Cu_3O_{6+x}$ are predominately two-dimensional (2D) in character. However, the finite coupling between CuO_2 bilayers results in a splitting of the spin excitations into acoustic and optical modes. The energy scale of the optical excitations is fairly large ($\omega \gtrsim 40$ meV) (Ref. 12) and has essentially no effect on the measurements presented below. The acoustic coupling results in a modulation of the scattered intensity along the 2D scattering rod $(\frac{1}{2},\frac{1}{2},l)$ (i.e., parallel to $\mathbf{c}^*$):

$$\mathcal{I}(\mathbf{Q},\omega)\sim\mathcal{S}(\mathbf{Q},\omega)=\sin^2(\pi l z)\mathcal{S}_{2d}(\mathbf{Q},\omega) .$$

Here $\mathcal{S}(\mathbf{Q},\omega)$ is the scattering function and $z=0.285$ is the distance between adjacent copper oxide planes in units of c. $\mathcal{S}_{2d}(\mathbf{Q},\omega)$ is only weakly dependent on momentum transfers parallel to $\mathbf{c}^*$. As the modulation along $\mathbf{c}^*$ is both energy and temperature independent, our measurements were done at $l=-1.8$ to maximize the measured intensity. (The sign choice takes advantage of the focusing properties of the spectrometer.)

III. EXPERIMENTAL RESULTS

In Fig. 1 we show the results of scans across the 2D antiferromagnetic zone center at 3 and 8 meV. The scattering intensity at 3 meV is clearly strongly suppressed with respect to the 8-meV data over the entire zone diagonal.[9] As shown by Tranquada *et al.*,[12] these data are readily fitted with a Gaussian line shape. The width of the scans across the 2D rod $(\frac{1}{2},\frac{1}{2},l)$ is, within error, independent of T and only weakly dependent on ω over the range of temperatures and energies studied here.[12,14] This is illustrated in Fig. 2, which shows the cross-rod scan width as a function of temperature for energy transfers of 5 and 12 meV. The resulting fullwidth at half maximum value of

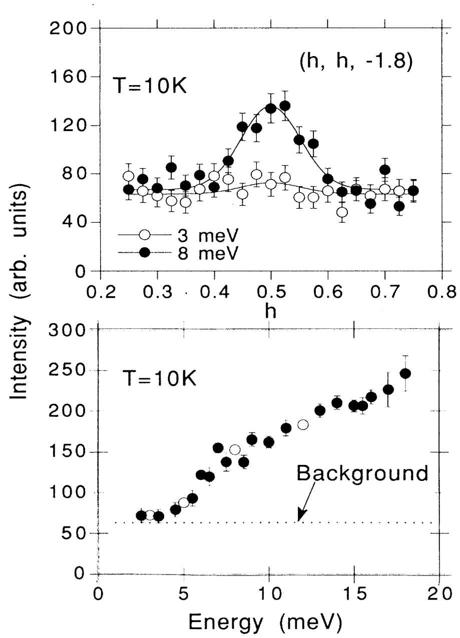

FIG. 1. (top) 3- and 8-meV scans across the 2D antiferromagnetic zone center $\mathbf{Q}_{\mathrm{AF}}\equiv(\frac{1}{2},\frac{1}{2},l)$ at $l=-1.8$. The scattering intensity at 3 meV is clearly strongly suppressed with respect to the 8-meV data over the entire zone diagonal. (bottom) Energy dependence of the peak intensity at the zone center from constant energy scans across the 2D zone center (○) and measurements at fixed $\mathbf{Q}_{\mathrm{AF}}\equiv(\frac{1}{2},\frac{1}{2},-1.8)$ (●). Below ~4 meV the difference between the measured intensity and the background level is, within error, zero.

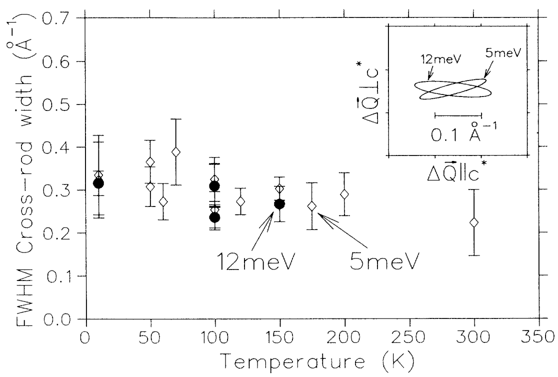

FIG. 2. Width of cross-rod scans vs temperature at 5 meV (◇) and 12 meV (●). (inset) Spectrometer resolution, in the scattering plane, parallel and perpendicular to the $\mathbf{c}^*$ axis.

~ 0.3 Å^{-1} corresponds to a length scale of $\xi\sim 2a$. In the lower panel of Fig. 1, we show the energy dependence of the peak intensities at 10 K resulting from scans across the 2D zone center (○) and measurements at fixed $\mathbf{Q}_{\mathrm{AF}}\equiv(\frac{1}{2},\frac{1}{2},-1.8)$ (●). Below 4 meV the difference between the measured intensity and the background level is, within error, zero. Above 4 meV the intensity increases gradually with increasing energy.

The imaginary part of the dynamic susceptibility is related to the scattering function $\mathcal{S}(\mathbf{Q},\omega)$ by the detailed balance factor

$$\mathcal{S}(\mathbf{Q},\omega)\propto[n(\omega)+1]\chi''(\mathbf{Q},\omega) .$$

For comparision with the results of other techniques and measurements on other systems, it is convenient to express our results in terms of the dynamic susceptibility integrated about $\mathbf{Q}_{\mathrm{AF}}$. However, as the width of our cross-rod data has only a negligible energy dependence, and the acoustic modulation along $\mathbf{c}^*$ is independent of energy and temperature, the $\mathbf{Q}$-integrated dynamic susceptibility is simply proportional to the peak intensity at the 2D zone center,

$$\chi''(\omega)=\int\chi''(\mathbf{Q},\omega)d\mathbf{Q}\propto\chi''(\mathbf{Q}_{\mathrm{AF}},\omega) .$$

Figure 3 shows the temperature dependence of $\chi''(\omega)$ at 5, 8, 12, and 16 meV. The data at both 12 and 16 meV show a smooth, monotonic increase in χ'' with decreasing temperature. In contrast, both the 5- and 8-meV data begin to falloff at temperatures above T_c. This gradual downturn in χ'' above T_c differs markedly from recent results[15,16] on superconducting $La_{2-x}Sr_xCuO_4$, where well-defined peaks in the low-energy susceptibility are observed at T_c. The suppression of the 5- and 8-meV scattering immediately below their respective peak temperatures suggests a continued diminution of the susceptibility with decreasing T, $\chi''(T\rightarrow 0)\rightarrow 0$, as would be expected for a system with an excitation gap $\omega_g>8$ meV. This behavior contrasts sharply with the neutron measurements of Rossat-Mignod *et al.* in $YBa_2Cu_3O_{6.6}$,[10] who report $\chi''(\mathbf{Q}_{\mathrm{AF}},\omega)$ at small ω going to zero at $T_c(=59$ K). At lower temperatures the 8-meV data, and

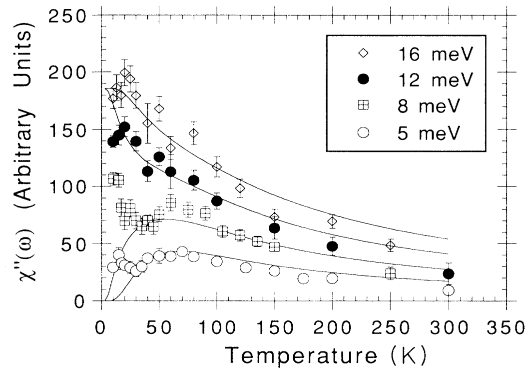

FIG. 3. Temperature dependence of χ'' at 5, 8, 12, and 16 meV. The solid lines are from a global fit to Eq. (1) (see text). The resulting gap energy is $\omega_g = 10(1)$ meV.

to a lesser extent the 5-meV data, show χ'' increasing with decreasing temperature. To verify this low-temperature behavior, additional 8-meV measurements were done using a fixed initial energy configuration, $E_i = 30.5$ meV, to check for possible contamination. As shown in Fig. 4, the value of χ'' resulting from the $E_i = 30.5$ meV and $E_f = 14.7$ meV data were found to agree within error. Qualitatively similar behavior in the temperature dependence of $\chi''(\omega, \mathbf{Q} = (0.7\pi, 0.7\pi))$ has recently been predicted by Zha, Levin, and Si (Fig. 2 in Ref. 17). However, these calculations result in a **Q** dependence of χ'' that is very much different from the roughly isotropic scattering we have observed about the antiferromagnetic zone center.[18]

The solid lines in Fig. 3 are the result of a global fit to the data using a function of the form

$$\chi''(\omega, T) = A \left[\tanh\left[\frac{(\omega - \omega_g)}{2T} \right] + \tanh\left[\frac{(\omega + \omega_g)}{2T} \right] \right] , \quad (1)$$

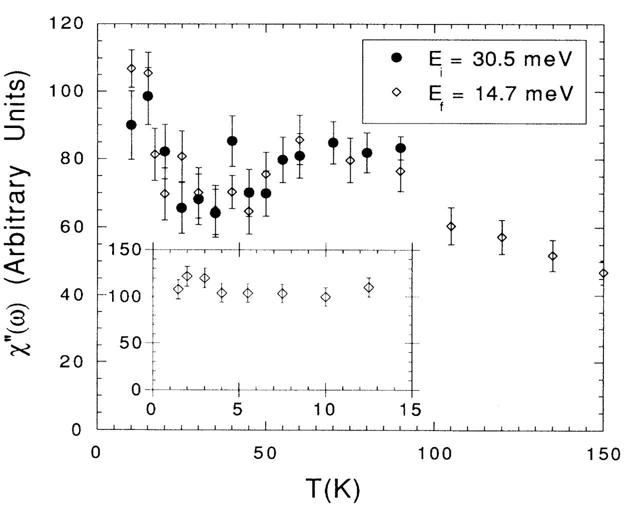

FIG. 4. Temperature dependence of χ'' at 8 meV measured both with fixed final energy $E_f = 14.7$ meV (◇) and fixed incident energy $E_i = 30.5$ meV (●) configurations. (inset) 8 meV χ'' at low temperature.

with temperature and energy independent amplitude A, and gap ω_g, parameters. As discussed in Ref. 12, Eq. (1) has no rigorous theoretical justification in the present application, though it is similar in form to the model of the imaginary part of the dynamic susceptibility at the antiferromagnetic zone center, $\chi''(\omega, \mathbf{Q}_{AF})$, developed by Bulut and Scalapino.[19] However, as demonstrated in Fig. 3, Eq. (1) does have the virtue of describing the gross features of χ'' at 16, 12, and (for $T \gtrsim 40$ K) 5 meV. The resulting value for the gap energy, $\omega_g = 10(1)$ meV, is consistent with the downturn in the 8-meV data below $T \sim 70$ K. The moderate success of this parameterization of $\chi''(\omega)$ in terms of a temperature-independent gap ω_g, suggests that the spin pseudogap observed in $YBa_2Cu_3O_{6+x}$ is not directly related to the onset of superconductivity. At lower temperatures, both the poor fit to the temperature dependence of the 8-meV data and the much smaller effective gap evident in the energy dependence of the scattering at 10 K (see Fig. 1) argue the need for a better parameterization of the functional form of χ''.

To illustrate better the dependence of the susceptibility on ω and T, we show χ'' vs T/ω in the top panel of Fig. 5. This plot clearly demonstrates that the temperature dependence of the susceptibility for a fixed energy transfer converges to a universal function of T/ω at high temperatures. This is in contrast to the scaling observed in $La_{2-x}Sr_xCuO_4$,[2] where an energy dependent amplitude normalization is needed to collapse the susceptibility at different energies into a single χ'' vs ω/T curve. Our data suggest that the temperature at which this universal behavior occurs increases with decreasing energy transfer. For instance, our 5-meV data appear to deviate from this generic behavior at temperatures below $T \sim 100$ K. This trend is consistent with the nearly elastic ($\omega \to 0$) NMR $1/T_1T$ results for the planar Cu sites in $YBa_2Cu_3O_{6.63}$ reported by Takigawa *et al.*[20] Again, the high temperatures at which these deviations occur argue that the mechanism responsible for the suppression of $\chi''(\omega)$ at low energies is not related to the onset of superconductivity. This is in strong contrast to the $La_{2-x}Sr_xCuO_4$ analysis of Mason, Aeppli, and Mook,[15] who suggest a direct relationship between the superconducting gap and spin excitation spectrum. To clarify the extent to which the Bose factor contributes to the overall scattering intensity, we also show the scattering function $\mathcal{S}(\omega)$ plotted on the same T/ω scale in the lower panel of the figure.

In Fig. 6 we show χ'' as a function of ω/T. We note that the data in this figure are qualitatively quite similar to the Fermi-liquid-based $\chi''(\mathbf{Q}_{AF}, \omega)$ calculations of Zha *et al.*[21] Again, the generic dependence of χ'' on ω/T at high temperatures (small ω/T) is clear. In contrast to the unusual low-temperature behavior of the 5- and 8-meV data discussed above, the 3-meV data for χ'' demonstrates a smooth decrease towards zero with falling temperatures (increasing ω/T). The low-temperature structure in the 8-meV susceptibility data, particularly the strong temperature dependence of χ'' at 10 K, is clearly seen in this figure. At low temperatures the data at both 12 and 16 meV have a nearly flat dependence on ω/T, though this behavior may be due in part to the exaggerated treatment of the low-temperature data with this coordinate choice.

The solid lines shown in Fig. 6 correspond to the fit to Eq. (1) discussed above. For comparision, we also show fits (dotted curves) of the 12- and 16-meV data using the functional form employed by Birgeneau *et al.*[11] in their scaling analysis of χ'' in $YBa_2Cu_3O_{6.5}$:

$$\chi''(\omega,T)=\chi''(\omega,0)\frac{2}{\pi}\tan^{-1}\left[\frac{\omega}{aT}\right], \qquad (2)$$

with $a=1.66$. These latter curves present a better fit to the low-temperature data, however, this is primarily due to the free amplitude parameters $\chi''(\omega,0)$. As mentioned

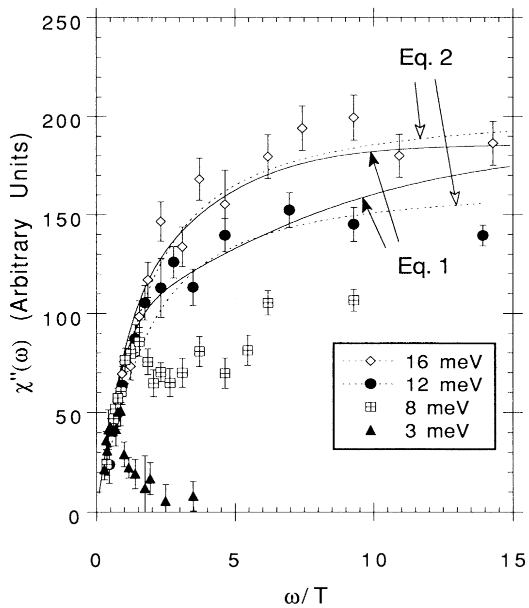

FIG. 6. χ'' vs ω/T. In contrast to the unusual low-temperature behavior of the 5- and 8-meV data (see text, Fig. 3), the 3-meV data shown here demonstrates a smooth decrease towards zero with decreasing energy. The solid (broken) lines are fits to the 12- and 16-meV data using Eq. 1 (Eq. 2) as described in the text.

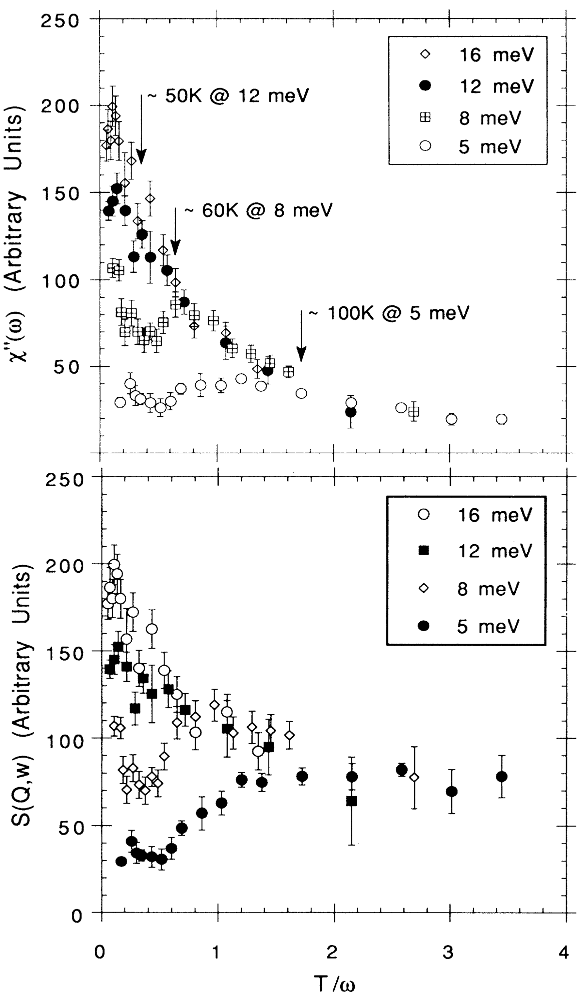

FIG. 5. (top) χ'' vs T/ω. The temperature dependence of the dynamic susceptibility for a fixed energy transfer converges to a universal function of T/ω at high temperatures. The arrows show that the temperature above which the generic behavior occurs increases with decreasing energy. (bottom) The corresponding scattering function $\mathcal{S}(\omega)$ vs T/ω.

in Ref. 11, the changing functional form of χ'' at lower energies due to the presence of a gap also limits the utility of a simple scaling approach in moderately and heavily doped $YBa_2Cu_3O_{6+x}$ compounds.

In conclusion, we have observed a universal dependence of $\chi''(\omega,T)$ on ω/T at high temperatures. The temperature above which this generic behavior is observed increases with decreasing energy in a fashion consistent with NMR results on $YBa_2Cu_3O_{6+x}$ samples with comparable transition temperatures.[20] These temperature-dependent measurements have not yet been extended to higher energies, where significant structure exists in the ω dependence of χ'' at lower temperatures.[12] Nonetheless, the generic ω/T dependence of our data at temperatures above $T\sim100$ K may account for the unusual normal-state behavior apparent in, for instance, resistivity[22] and optical conductivity[23] measurement of $YBa_2Cu_3O_{6+x}$. Above $T\sim40$ K our data is in large part well described by Eq. (1). The temperature independence of the pseudogap used in this model, and to a lesser extent, the small magnitude of the resulting gap energy, would argue that the pseudogap is not directly related to the onset of superconductivity. Perhaps the most surprising result of our measurements is the observation of a recovery in the scattering intensity at 5 and 8 meV at temperatures below $T\lesssim40$ K. The presence of this additional structure in χ'' at low temperatures clearly demonstrates the need for more detailed theoretical descriptions of χ''. In particular, the physics responsible for the suppression of χ'' at low energies/temperatures and its relation to superconductivity in $YBa_3Cu_3O_{6+x}$ require further clarification.

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