The spin anisotropy of the magnetic excitations in the normal and superconducting states of optimally doped YBa$_2$Cu$_3$O$_{6.9}$ studied by polarized neutron spectroscopy

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We use inelastic neutron scattering with spin polarization analysis to study the magnetic excitations in the normal and superconducting states of YBa$_2$Cu$_3$O$_{6.9}$. Polarization analysis allows us to determine the spin polarization of the magnetic excitations and to separate them from phonon scattering. In the normal state, we find unambiguous evidence of magnetic excitations over the 10–60 meV range of the experiment with little polarization dependence to the excitations. In the superconducting state, the magnetic response is enhanced near the “resonance energy” and above. At lower energies, $10 \lesssim E \lesssim 30$ meV, the local susceptibility becomes anisotropic, with the excitations polarized along the c-axis being suppressed. We find evidence for a new diffuse anisotropic response polarized perpendicular to the c-axis which may carry significant spectral weight.

I. INTRODUCTION

High temperature superconductivity (HTS) arises when certain two dimensional antiferromagnetic Mott insulators are electron or hole doped [1]. The antiferromagnetic parent compounds such as La$_2$CuO$_4$ show spinwave excitations up to $2J \approx 300$ meV [2]. Doping causes the magnetic response to evolve from that of spin waves to a more structured response [3–13], with strong spin fluctuations being observed for superconducting compositions in a number of systems including YBa$_2$Cu$_3$O$_{6+x}$ (YBCO) [7–10, 14, 15], La$_{2-x}$Sr$_x$CuO$_4$ [11, 16] and Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ [12, 13, 17]. Many optimally-doped cuprates show a strong well-defined collective magnetic excitation which is localised in reciprocal space and strongest near the $Q=(1/2,1/2)\equiv(\pi,\pi)$ position. It is sharp in energy and develops on cooling through the critical temperature. This excitation has become known as the “magnetic resonance”. The magnetic resonance has been observed in YBa$_2$Cu$_3$O$_{6+x}$ [4–6], Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ [17], Tl$_2$Ba$_2$CuO$_{6+\delta}$ [18] and HgBa$_2$CuO$_{4+\delta}$ [19].

The magnetic resonance is certainly the strongest feature in the magnetic excitations spectrum of the materials listed above, however, it only accounts for a small fraction (≈2%) [9, 15, 17] of the total scattering expected from the unpaired $3d$ electrons of the Cu atoms. In this work we search for other contributions to the response which are spread out in energy and wavevector but nevertheless may carry significant spectral weight. These are harder to observe because they are weak and may not show the strong temperature dependence which allows the resonance to be easily isolated. We use inelastic neutron scattering with polarization analysis to isolate the magnetic scattering from phonon scattering.

We find that there is a significant response in the normal state which can account for much of the spectral weight from which the resonance is formed. In the superconducting state, we find evidence for a diffuse contribution at energies well below the resonance. This new contribution is polarized with strong fluctuations perpendicular to the c-axis.

II. BACKGROUND

A. Polarization Analysis

Neutrons scatter from condensed matter via two processes: (i) The electromagnetic interaction probes fluctuations in the magnetization density of the electrons (in this paper this is referred to as magnetic scattering). (ii) The strong nuclear force is responsible for scattering from the atomic nuclei. The nuclear scattering allows us to probe phonons which are correlations (in time and space) between the position of the nuclei. The existence of two distinct scattering processes makes the neutron an extremely versatile probe. However, it also means that the two types of scattering can mask each other.

Polarization analysis of the neutron’s spin allows the separation of magnetic and nuclear (phonon) scattering. In the present work, we use longitudinal polarization analysis (LPA). In LPA, a spin-polarized incident neutron beam is created and its polarization maintained by a small magnetic field (~1 mT). The number of neutrons scattered with spins parallel or antiparallel to this quantizing field are then measured. We label each spin-polarization state as parallel ($\equiv\text{up,}\uparrow\downarrow$) or antiparallel ($\equiv\text{down,}\downarrow\uparrow$) to the applied field. The cross sections are referred to as spin-flip (SF) ($\uparrow\rightarrow\downarrow,\downarrow\rightarrow\uparrow$) or non-spin-flip (NSF) ($\uparrow\rightarrow\uparrow,\downarrow\rightarrow\downarrow$). A natural reference frame in which to understand the cross sections is one referenced to the scattering vector $\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$ of the neutron, where $\mathbf{k}_i$ and $\mathbf{k}_f$ are the incident and final wavevectors of the neutron. Thus, $\hat{\mathbf{x}} \parallel \mathbf{Q}$, $\hat{\mathbf{y}} \perp \mathbf{Q}$, and $\hat{\mathbf{z}} \perp \mathbf{Q}$ and $\perp$ to the spectrometer scattering plane (the plane containing $\mathbf{k}_i$ and $\mathbf{k}_f$). We make measurements with the neutrons

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polarized along each of these axes.

The neutron cross sections as a function of spin polarization have been derived and presented elsewhere [20–24]. The spin-flip magnetic cross section for spin polarization $\parallel Q$ is

$$\sigma_{xx}^{\uparrow\downarrow} = \left( \frac{d^2\sigma}{d\Omega dE} \right)_{H||x} = k_f (\gamma r_e)^2 \frac{1}{h_i^2 \mu_B^2 \pi} F^2(Q) \chi''_{yy}(q, \hbar \omega) + \chi''_{zz}(q, \hbar \omega) \frac{1}{1 - \exp(-\hbar \omega/kT)},$$

where $(\gamma r_e)^2=0.2905$ barn sr$^{-1}$ and $|F(Q)|^2$ is the anisotropic magnetic form factor for a Cu$^{2+}$ $d_{x^2-y^2}$ orbital. $\chi''_{rr}(q, \hbar \omega)$ is the generalized susceptibility corresponding to magnetic fluctuations along the $\nu$-axis. Thus, for example:

$$\langle m^2_r(q, \omega) \rangle = \frac{1}{\pi} \frac{\chi''_{xx}(q, \omega)}{1 - \exp(-\hbar \omega/kT)},$$

where the angle brackets denote thermal averages. The spin-dependent cross sections including the nuclear incoherent cross sections (i.e. the phonon cross section) $N(q, \omega)$ are:

$$\sigma_{xx}^{\uparrow\downarrow} \propto \chi''_{yy}(q, \omega) + \chi''_{zz}(q, \omega) + BG_{\uparrow\downarrow},$$
$$\sigma_{yy}^{\uparrow\downarrow} \propto \chi''_{zz}(q, \omega) + BG_{\uparrow\downarrow},$$
$$\sigma_{zz}^{\uparrow\downarrow} \propto \chi''_{yy}(q, \omega) + BG_{\uparrow\downarrow},$$
$$\sigma_{xx}^{\uparrow\downarrow} \propto N(q, \omega) + BG_{\uparrow\uparrow},$$
$$\sigma_{yy}^{\uparrow\downarrow} \propto \chi''_{yy}(q, \omega) + N(q, \omega) + BG_{\uparrow\downarrow},$$
$$\sigma_{zz}^{\uparrow\downarrow} \propto \chi''_{zz}(q, \omega) + N(q, \omega) + BG_{\uparrow\uparrow},$$

where we have neglected the nuclear spin incoherent cross-section which is small in the present experiments [25] and BG denotes the background for the configuration. In this work we isolate two components of the susceptibility by comparing different SF cross sections:

$$\sigma_{xx}^{\uparrow\downarrow} - \sigma_{yy}^{\uparrow\downarrow} \propto \chi''_{yy}(q, \omega)$$
$$\sigma_{xx}^{\uparrow\downarrow} - \sigma_{zz}^{\uparrow\downarrow} \propto \chi''_{zz}(q, \omega)$$

**B. Bilayer Effects**

YBa$_2$Cu$_3$O$_{6+x}$ has two CuO$_2$ planes per unit cell (See Fig. 1). The usual starting point for models of the magnetic response is to neglect the electronic coupling between CuO$_2$ planes in different unit cells and include only coupling between the CuO$_2$ planes of the bilayer located in the center of the unit cell in Fig. 1. This leads to a pair of bonding (b) and antibonding (a) energy bands.

The presence of a mirror plane between the two planes of the bilayer means that the magnetic excitations have distinct odd (o) or even (e) character. In this description, the magnetic response is of the form [26–29]

$$\chi''(h, k, l, \omega) = \chi''_{e}(h, k, \omega) \cos^2 \left( \frac{\pi d}{c} \right)$$
$$+ \chi''_{o}(h, k, \omega) \sin^2 \left( \frac{\pi d}{c} \right),$$

where $d$ is the separation of the CuO$_2$ planes. For YBa$_2$Cu$_3$O$_{6.9}$ $d=3.38$ Å, this means the odd response is strongest at $l=(n+1/2)c/2d=1.73, 5.3, \ldots$ The strongest features in the magnetic response of YBa$_2$Cu$_3$O$_{6+x}$ observed by INS are in the odd channel [4–6] and we measure the odd channel in the present experiment. We note that weaker resonance features have been reported in the even channel [30, 31] for various dopings. The reported even resonance occurs at higher energy than in the odd channel.

**C. Sample Details**

We investigated a near optimally doped sample of YBa$_2$Cu$_3$O$_{6.9}$ ($T_c=93$ K) grown by a top seed melt growth technique [32]. YBa$_2$Cu$_3$O$_{6.9}$ has the ortho-I structure show in Fig. 1 with lattice parameters $a=3.82$ Å, $b=3.89$ Å and $c=11.68$ Å ($T=77$ K) [33]. The single crystal studied in the present experiment is twinned and the results presented are an average over the two twin domains. The crystal had a mass of 32.5 g and mosaic spread 1.3''. It was annealed for 17 days at 550°C, followed by 13 days at 525°C, in oxygen to achieve the required oxygen stoichiometry. Neutron depolarization measurements (see Fig. 4) indicated that $T_c$(onset) = 93±0.2 K. Based on $T_c$ and the heat treatment [34, 35], we estimate the oxygen stoichiometry to
be $x=0.9 \pm 0.01$.

D. Experimental Setup

Experiments were performed using the IN20 three-axis spectrometer at the Institut Laue-Langevin, Grenoble using a standard longitudinal polarization analysis set up. Neutron polarization analysis was carried out using a focusing Heusler monochromator and analyzer. The sample was mounted with the [310] and [001] directions in the horizontal scattering plane of the instrument. We worked using a sample goniometer to access reciprocal space and to the scattering vector $Q$ which meant that the neutron polarizations and hence the measured susceptibilities are not along the crystallographic axes (see Fig. 2). For example, the angle between the $y$-axis and the crystallographic $c$-axis is $\theta=20.6^\circ$. This leads to a small mixing of the different components of the susceptibility during the measurement. Thus:

$$
\sigma_{\text{corr}}^{\text{meas}} = \frac{F}{F-1} \sigma_{\text{SF}}^{\text{meas}} - \frac{1}{F-1} \sigma_{\text{NSF}}^{\text{meas}},
$$

where the flipping ratio $F \approx 7.5$ was determined from measurements on Bragg peaks made under the same conditions. For experimental reasons, measurements were made with neutrons polarized parallel and perpendicular to the scattering vector $Q$ and to the scattering vector $Q$. Neutrons are polarized along the $\hat{x}$, $\hat{y}$ or $\hat{z}$ axes. $\hat{x}$ is parallel to $Q$, $\hat{z}$ is perpendicular to $Q$ and in the $(h,k,0)$ plane. Thus $\hat{x} \parallel (1.5a^*+0.5b^*+1.8c^*)$, $\hat{y} \parallel (0.5a^*+0.18b^*+4.6c^*)$, $\hat{z} \parallel (-0.5a^*+1.5b^*)$, $\theta = 20.6^\circ$ and $\cos^2 \theta = 0.88$. (b) Illustration of the area in reciprocal space where the measurements in Sec. III B were made. For $E < 52$ meV, we used data collected over the black square ($1.3 \leq h \leq 1.5$ and $0.3 \leq k \leq 0.5$) to infer $\chi''(\omega)$ measured over the grey area. Data in Fig. 6 covers the black square plus dotted area.

Fig. 2. (Color online) (a) Illustration of the reference frame used to describe polarization analysis. Neutrons are polarized along the $\hat{x}$, $\hat{y}$ or $\hat{z}$ axes. $\hat{x}$ is parallel to $Q$, $\hat{z}$ is perpendicular to $Q$ and in the $(h,k,0)$ plane. Thus $\hat{x} \parallel (1.5a^*+0.5b^*+1.8c^*)$, $\hat{y} \parallel (0.5a^*+0.18b^*+4.6c^*)$, $\hat{z} \parallel (-0.5a^*+1.5b^*)$, $\theta = 20.6^\circ$ and $\cos^2 \theta = 0.88$. (b) Illustration of the area in reciprocal space where the measurements in Sec. III B were made. For $E < 52$ meV, we used data collected over the black square ($1.3 \leq h \leq 1.5$ and $0.3 \leq k \leq 0.5$) to infer $\chi''(\omega)$ measured over the grey area. Data in Fig. 6 covers the black square plus dotted area.

III. RESULTS

A. Energy- and Wavevector-Dependent Scans

Fig. 3 shows energy-dependent scans made at the $(1.5,0.5,1.73)$ position with various spin polarizations. At this position in reciprocal space the non spin-flip (phonon) scattering is up to 8 times stronger than the spin-flip scattering. Thus an unpolarized measurement made under the same conditions would be dominated by phonon scattering at some energies (the comparison with unpolarized experiments is discussed further in Appendix B). In the normal state the $\sigma_{xx}^{\uparrow\downarrow}$ cross section is larger than $\sigma_{yy}^{\uparrow\downarrow}$ and $\sigma_{zz}^{\uparrow\downarrow}$ over a wide energy range, $20 \lesssim E \lesssim 60$ meV, signalling the presence of magnetic
excitations. We can use Eq. 8 to isolate the out-of-plane response \( \chi_c'' \); this is shown in Fig. 3(c). In the superconducting state there is a large increase in \( \sigma_{xx}^{\uparrow \downarrow} \) and \( \sigma_{yy}^{\uparrow \downarrow} \) (\( \sigma_{zz}^{\uparrow \downarrow} \) was not measured in this case) near the resonance energy. The difference scan Fig. 3(c) shows a sharp resonance peak at \( E \approx 40 \) meV which appears to have formed by a transfer of spectral weight from lower energies \( E \lesssim 35 \) meV. The \( \chi_c'' \) response appears to be largely gapped below about 30 meV. Similar data was obtained using unpolarized neutrons by Bourges et al. [37]. We do not observe a collective magnetic excitation in the 50–60 meV range as observed recently in HgBa\(_2\)CuO\(_{4+\delta}\) [38]. We note that there is a peak in the non spin flip channel in this energy range in Fig. 3(a).

In order to analyze our data further, we fitted the \( T = 10 \) K scan in Fig. 3(c) to the resolution-corrected model cross section

\[
\chi_c(q, \omega) = [A\delta(\omega - \omega_0) + B\delta(\omega - \omega_0)] \\
\times \exp \left\{ -\frac{(h - 1/2)^2 + (k - 1/2)^2}{2\sigma^2} \right\},
\]

where \( \theta \) is the heaviside step function and \( \sigma \) is the width parameter extracted from a \( q \)-dependent scan through the resonance (see Table I). Throughout this paper we use the RESTAX simulation package [39] to perform convolutions of the instrumental resolution function and model cross sections. Using the cross section defined by Eq. 9, we find that the width of the peak due to the resonance in Fig. 3(c) is resolution limited and \( \hbar \omega_0 = 41 \pm 1 \) meV.

We have converted the data in Fig. 3(c) to absolute units using Eq. 1 without attempting to deconvolve the experimental resolution. This means that each point in the scan is an average (in wavevector and energy) over the instrumental resolution. Keeping this in mind, we have integrated the response in Fig. 3(c) in energy for

\[
\chi_c(Q, \omega) = \int\int \chi_c(q, \omega) \sin(Qr) dq \, dq \, d\omega.
\]
In the normal state, there is a magnetic response at all three energies. On cooling to \( T = 10 \text{ K} \), the lower frequency \( E = 26 \text{ meV} \) response is suppressed while the response at the resonance energy \( (E = 40 \text{ meV}) \) increases dramatically and the \( q \)-width decreases. The data were fitted to a model consisting of four incommensurate peaks with locations \( \mathbf{Q}_d = (1/2 \pm \delta, 1/2) \) and \( (1/2, 1/2 \pm \delta) \) and width \( \sigma \):

\[
\chi''(\mathbf{q}, \omega) = A \sum \exp \left\{ -\frac{(\mathbf{Q} - \mathbf{Q}_d)^2}{2\sigma^2} \right\}. \tag{10}
\]

The results of this fitting procedure are shown in Table I.

We first consider the scans at the resonance energy \( (\hbar \omega = 40 \text{ meV}) \). A single Gaussian peak \( (\delta = 0) \) provides a good description of the scan in the superconducting state [Fig. 4(a) and Fig. 5(a)]. In the normal state, there is magnetic scattering at the resonance energy [Fig. 5(b)]. The existence of a magnetic response at this energy in optimally doped YBCO has been a subject of some debate [4–6, 37, 40] and we will discuss this later. It is clear from our data that the response at the resonance energy is broader in \( q \) and weaker in the normal state than the superconducting state. If we fit the 40 meV data using Eq. 10 with \( \delta = 0 \), we find \( \sigma = 0.18 \pm 0.02 \) and 0.115 \pm 0.01 for the normal and superconducting states respectively. Returning to the superconducting state data at lower energy, we find a single Gaussian peak \( (\delta = 0) \) does not provide a good description of the \( \hbar \omega = 34 \text{ meV} \) \( (T = 10 \text{ K}) \) scans [Fig. 4(c) and Fig. 5(c)] in the superconducting state. Better fits are obtained when a finite incommensurability \( \delta = 0.12 \pm 0.02 \) is used. This \( \delta \) is in agreement with that obtained in other studies of optimally doped YBCO [15, 40]. In the normal state [Fig. 4(b,d,f) and Fig. 5(b,d,f)] we see clear magnetic scattering at the three energies investigated. We do not see a two-peak structure as in Fig. 4(c), instead the response appears to be broadened out into single peak which, in some cases [e.g. Fig. 4(b,f)], looks “flat topped”. To contrast the normal and superconducting state responses, we have fitted the scans with the value of \( \delta \) determined from the \( T = 10 \text{ K} \) and \( \hbar \omega = 34 \text{ meV} \) scan. The normal state response is broader in all cases (see Table I).

### B. Local Susceptibility Measurements

In order to search for the diffuse contributions to the magnetic response, we sampled a grid of points near the \((3/2,1/2)\) position where the response is generally stronger. Extended grids at two characteristic energies are shown in Fig. 6. For this part of the experiment we collected three spin-flip channels and we were able to extract \( \chi''_{ab} / \hbar \) and \( \chi''_{c} \). The lowest row of Fig. 6 shows the signal extracted via Eq. 4. The data collected at \( E = 40 \text{ meV} \) shows that the response is strongest near the \((1.5,0.5,1.73)\) position both in the normal and superconducting states. At \( E = 26 \text{ meV} \), we see a normal state response which is spread out; see, for example, \( \chi''_{ab} / \hbar (E = 26 \text{ meV}, T = 100 \text{ K}) \), where the upper part of

| \( T \)(K) | \( \hbar \omega \)(meV) | \( \delta \)(r.l.u.) | \( \sigma \)(r.l.u.) |
|--------|-----------------|-----------------|-----------------|
| 10     | 26              | N/A             | N/A             |
| 34     | 0.12 ± 0.02     | 0.059 ± 0.01    |
| 40     | 0               | 0.114 ± 0.01    |
| 94     | 26              | 0.12 ± 0.05     |
| 34     | 0.12 ± 0.095    | 0.01           |
| 40     | 0.12 ± 0.071    | 0.01           |
| 40     | 0               | 0.16 ± 0.02     |

TABLE I. Incommensurability \( \delta \) and width \( \sigma \) parameters obtained from fitting Eq. 10 to the scans in Fig. 4. Where no error is quoted, the parameter was fixed.
the map shows signal. On entering the superconducting state $\chi''$ shows a much larger change than $\chi''_{ab}$ suggesting that a spin anisotropy develops.

Fig. 7 shows the wavevector integrals collected at a number of energies over the grey region shown in Fig. 2. This is the region of highest intensity in the Brillouin zone, but there is clearly scattering in other parts of the zone. The contribution of the grey region to $\chi''_c(\omega)$ is shown in Fig. 7(c) and (d). Fig. 7 shows that there is a strong response in the normal state over a wide energy range. When compared to the energy-dependent scan at (1.5,0.5,1.73), we see that the higher energy response is relatively stronger. This is due to the presence of a broader response in $q$ at higher energies $E \gtrsim 50$ meV \cite{10, 15, 41}. On entering the superconducting state, we see a strong reduction in $\chi''_c$ with little change in $\chi''_{ab}$. This means the magnetic response develops a strong spin anisotropy in the superconducting state (see Sec. IV B for more discussion). For higher energies, $E \geq 34$ meV, the response increases in the superconducting state, not only at the resonance energy, but up to 60 meV. Table II shows that when integrated over the range $12 < E < 60$ meV the total fluctuating moment $\langle m^2 \rangle$ increases by about 60%. In order to compare with other studies of the resonance in near optimally doped YBCO \cite{9, 15, 17}, we have also integrated the data in Fig. 7 over the smaller energy range $30 < E < 60$ meV (see Table II) in this case we see a larger change in $\langle m^2 \rangle$ (between the normal and
TABLE II. Fluctuating moments $\langle m_n^2 \rangle$, $\langle m_z^2 \rangle$ and $\langle m^2 \rangle = 2(\langle m_n^2 \rangle + \langle m_z^2 \rangle)$ in the normal ($T = 100$ K) and superconducting ($T = 10$ K) states calculated by numerically integrating the response in Fig. 7. The errors quoted are statistical and do not include the systematic error in the absolute scale which is about $\pm 20\%$.

| $T$(K) | $\langle m_n^2 \rangle/(\mu_B^2 \text{ f.u.}^{-1})$ | $\langle m_z^2 \rangle/(\mu_B^2 \text{ f.u.}^{-1})$ | $\langle m^2 \rangle/(\mu_B^2 \text{ f.u.}^{-1})$ |
|--------|-----------------|-----------------|-----------------|
| 12     | $\leq \hbar\omega$(meV) $\leq 60$ | $0.031 \pm 0.004$ | $0.026 \pm 0.004$ | $0.088 \pm 0.007$ |
| 100    | $30 \leq \hbar\omega$(meV) $\leq 60$ | $0.017 \pm 0.003$ | $0.022 \pm 0.003$ | $0.056 \pm 0.005$ |
| 10     | $0.024 \pm 0.003$ | $0.026 \pm 0.003$ | $0.074 \pm 0.005$ |
| 100    | $0.009 \pm 0.002$ | $0.014 \pm 0.002$ | $0.032 \pm 0.003$ |

FIG. 8. Schematic representation of $\chi''(\mathbf{q}, \omega)$ in the superconducting state of YBa$_2$Cu$_3$O$_{6.9}$ based on Refs. 42 and 43. The black line is the resonance mode and grey area the particle-hole continuum. Scans (a), (b) and (c) correspond approximately to 20, 40 and 60 meV.

IV. DISCUSSION

A. Response in the Normal and Superconducting States

Theories of the magnetic excitations in the superconducting state of cuprate superconductors such as YBa$_2$Cu$_3$O$_{6.9}$ are well developed [42–53]. Many features are explained by a magnetic exciton scenario [42, 43, 46, 47, 52] in which the resonance is a bound state in the particle-hole channel, which appears in a region of $\mathbf{q} - \omega$ space where there are no damping processes due to electron-hole pair creation. This is illustrated schematically in Fig. 8. In such a picture, significant magnetic response should also be present in the normal state. As the system enters the superconducting state we expect the low energy response to be suppressed below $E \lesssim \Delta$ and an enhancement of the response at the resonance energy. This is the behaviour seen in Figs. 3 and 5. The nature of the magnetic response in the normal state of optimally doped YBCO has been a subject of debate, particularly with regard to energies near the resonance energy [4–6, 37, 40, 54]. Some studies suggest there is a significant response [4, 37] for $\mathbf{q} \approx (1/2, 1/2)$ and $\hbar\omega \approx 40$ meV, while others claim the response is absent or too weak to observe [6, 40, 54]. The present experiment allows the magnetic response to be separated from phonon scattering. We find that the out-of-plane response $\chi''(\mathbf{q}, \omega)$ is peaked around $\hbar\omega \approx 30$ meV for $\mathbf{q} \approx (1/2, 1/2)$ in the normal state ($T = 94$ K). On cooling there is a shift of spectral weight to higher energies which leads to the formation of the resonance peak near 40 meV, with the concomitant formation of incommensurate peaks observed at 34 meV and a spin gap below about 30 meV for the $\chi''$ component of the response. This is consistent with the formation of a magnetic excitonic mode as illustrated schematically in Fig. 8. The work presented in this paper refers to optimally doped YBCO where it is harder to separate the magnetic contribution from phonons and other background scattering than for other compositions. We note that for underdoped YBCO (e.g. YBa$_2$Cu$_3$O$_{6.6}$) [8, 10, 55, 56] a strong dispersive excitonic mode is also observed in the superconducting state. On warming to $T_c$ the remnants of this mode are clearly observable and persist well above $T_c$.

The discussion above relates to the energy- and wavevector- dependent scans presented in Sec. IIIA. These yield information about the out-of-plane fluctuations described by $\chi''$. We did not collect the corresponding scans for $\chi''/\chi''$, however, we did probe this component of the local susceptibility in the measurements presented in Sec. IIIB. These measurements were designed to yield estimates for the total response in a region of q space rather than identify the location of specific features such as incommensurate peaks. They are summarized in Fig. 7(c) and (d). In Fig. 7(c) we see that there is strong evidence for additional scattering below 30 meV in the $\chi''/\chi''$ component of the response. This response appears to be rather spread out in wavevector when we inspect the corresponding map ($\hbar\omega = 26$ meV, $T = 10$ K) in Fig. 6. Thus our results suggest that there are other (diffuse) contributions to the $\chi''/\chi''$ response at low energies in the superconducting state. The $\chi''/\chi''$ component of the response has a lower ‘spin gap’ than the $\chi''$ component. The low energy response ($E \lesssim 30$ meV) may be due to the electron-hole continuum also present in the theories of the resonance [42, 43, 52]. This is illustrated schematically in Fig. 8.

B. Spin Anisotropy in YBa$_2$Cu$_3$O$_{6.9}$

Our results suggest that a spin anisotropy develops in the lower energy ($10 \lesssim E \lesssim 30$ meV) excitations on entering the superconducting state. Nuclear magnetic resonance
(NMR) probes the spin fluctuations in the very low frequency limit and, indeed, the anisotropy of spin-lattice relaxation rate \( T_1 \) in YBa\(_2\)Cu\(_3\)O\(_y\) has been reported to show a strong temperature dependence in the superconducting state [57, 58]. Various theories have attributed this to the combined effect of the NMR form factor and a changing \( \chi''(q, \omega) \) (See e.g. Ref. 59 and 60). However, the present measurements show that there is also an significant intrinsic anisotropy in \( \chi''(q, \omega) \) with respect to the spin direction which must be considered. It is interesting to note that Uldry et al. [61] have extracted the intrinsic anisotropy from NMR data and concluded that the out-of-plane correlations do not change appreciably on entering the superconducting state, in contrast to our results. This may be because NMR measurements probe the excitations at much lower frequencies than our measurements.

Anisotropy in the susceptibility ultimately comes from the spin-orbit interaction. An exotic case is the superfluid 3He A-phase [62], where the susceptibility depends on the orientation of the angle of the field to the characteristic spin vector \( \mathbf{d} \). In the case of superconductors, dramatic changes in a pre-existing spin anisotropy have recently been observed in BaFe\(_{1.5}\)Ni\(_{0.1}\)As\(_2\) [36] and a small anisotropy at the resonance energy is observed in FeSe\(_{0.5}\)Te\(_{0.5} \) [63]. A possible origin of the emergence of spin anisotropy in YBa\(_2\)Cu\(_3\)O\(_y\) may be the Dzyaloshinskii-Moriya (DM) interactions between the copper spins [64]. The buckled structure of the CuO\(_2\) planes in ortho-I YBa\(_2\)Cu\(_3\)O\(_y\) (see Fig. 1) means that DM interactions of the form \( \mathbf{D} \cdot \mathbf{S}_i \times \mathbf{S}_j \) are allowed between neighbouring Cu spins. The presence of such terms leads to additional spin anisotropy. This leads to a polarization dependence to the spin wave dispersion and energy in the antiferromagnetic parent compounds La\(_2\)CuO\(_4\) [65] and YBa\(_2\)Cu\(_3\)O\(_{6.2}\) [66]. In the case of YBa\(_2\)Cu\(_3\)O\(_{6.2}\) the anisotropy gaps are \( \sim 10 \) meV [66] and the ordered moments lies along the [100] direction [67].

The low energy excitations (\( E \lesssim 30 \) meV) we observe have their predominant fluctuations within the CuO\(_2\) planes making the \( a/b \) response largest. At higher energies, \( E \approx 40 \) meV, the excitations are more isotropic. This corresponds to all three components of the spin-triplet \( \{ | ↑↑⟩, | ↑↓⟩− | ↓↑⟩, | ↓↓⟩ \} \) being excited.

### Appendix A: Sum Rules and the Magnetic Response

#### 1. Local Susceptibility

The local susceptibility is a useful way to characterise the overall response. It is defined as,

\[
\chi''(\omega) = \int \frac{\chi''(Q, \omega) d^3Q}{\int d^3Q}, \tag{A1}
\]

where, in general, the integrals are over a volume of reciprocal space which samples the full \( Q \) dependence of \( \chi''(Q, \omega) \). In the case of YBa\(_2\)Cu\(_3\)O\(_{6+x}\) this is one unit cell in the \( ab \) plane and infinity along \( c \). The local susceptibility can be split into the two terms of Eq. 5. Thus integrating Eq. 5 we have

\[
\chi''(\omega) = \chi''_0(\omega) + \chi''_1(\omega), \tag{A2}
\]

where

\[
\chi''_0(\omega) = \frac{1}{2} \int_0^1 dh \int_0^1 dk \chi''_0(h, k, \omega). \tag{A3}
\]

The definition for \( \chi''_0(\omega) \) used here differs by a factor 2 from earlier work, but allows a direct comparison with single layer compounds [11].

#### 2. Total Moment Sum Rule

For an ion with spin only moment, the total squared moment is

\[
\langle m^2 \rangle = g^2 \mu_B^2 S(S + 1) = 3 \mu_B^2 \text{ for } S = \frac{1}{2} \text{ and } g = 2. \tag{A4}
\]

The total fluctuating moment observed by INS over a given range of energy and momentum can be determined from the fluctuation-dissipation theorem and is

\[
\langle m^2 \rangle = \langle m_x^2 + m_y^2 + m_z^2 \rangle = \frac{1}{\pi} \int \left[ \frac{\chi''_{xx}(\omega) + \chi''_{yy}(\omega) + \chi''_{zz}(\omega)}{1 - \exp(-\hbar \omega/kT)} \right] d\omega. \tag{A5}
\]
Appendix B: Comparison with Unpolarized Studies

There are many unpolarized studies of the magnetic excitations in YBa$_2$Cu$_3$O$_{6+x}$ [4, 6, 14, 40, 41]. In this section we show that our results are broadly consistent with previous results. The main issues that arise in unpolarized studies are: (i) the separation of magnetic signal from background and (ii) the separation of magnetic and phonon scattering. In the present spin-polarized study we may compare to different spin-flip cross-sections to remove the background and the phonon contribution. This is demonstrated in Eqs. 3-4.

The unpolarized inelastic cross section is generally of the form

$$\left(\frac{d^2\sigma}{d\Omega dE}\right) \propto \frac{\chi''(q, \omega, T)}{1 - \exp(-\hbar\omega/kT)} + N(q, \omega, T), \quad \text{(B1)}$$

where the first term represents the inelastic magnetic response and the second that due to the phonons. A sharp magnetic response such as the resonance can be isolated through $q$ and $\omega$ scans and verified as being magnetic through the form factor present in Eq. 1. However, a broad or diffuse response is more difficult to distinguish from phonons. The phonon response $N(q, \omega, T)$ usually decreases with temperature ($\hbar\omega \lesssim kT$) or remains constant ($\hbar\omega \gg kT$) due to the Bose factor. Thus a signal that increases with decreasing temperature (such as the resonance) is likely to be magnetic. If a magnetic signal decreases with decreasing temperature such as the response below about 30 meV in Fig. 3(c) it is difficult to distinguish from phonons using unpolarized neutrons.

In Figs. 9 and 10, we have reconstructed ‘unpolarized’ scans by adding together the spin-flip and non-spin-flip intensities for $H \parallel x$, $\sigma_{xx}^{\uparrow\downarrow} + \sigma_{xx}^{\downarrow\uparrow}$. Our experiment was not optimized for this reconstruction because the spin-flip channels were counted longer than non-spin-flip, nevertheless we can make some useful observations. As expected, Fig. 9(a) clearly shows the resonance at $T = 10$ K and $E = 40$ meV in the superconducting state. Note there is increased background or phonon scattering at larger $k$ in this scan. In the normal state, at $T = 94$ K, it is not possible to identify any magnetic scattering. For $E = 34$ meV [Fig. 9(b)], the scans at both temperatures are similar. The data are consistent with a broad magnetic response which changes little between the two temperatures [see Fig. 5(c)-(d)]. Finally, for $E = 26$ meV we observe a decrease in intensity across much of the scan on lowering the temperature. This is consistent with a reduction of the magnetic response at this energy [see Fig. 5(e)-(f)]. However, the phonon scattering at this energy and wavevector is strong [see Fig. 3(a)] thus part (about 50%) the reduction observed using unpolarized spectroscopy is due to the change of the Bose factor for
the phonons.

Fig. 10 shows energy-dependent scans at the $Q=(1.5,0.5,1.73)$ position and a temperature difference often used to isolate the resonance (see e.g. [6, 17]). From Figs. 3 and 4, we can deduce that about 50% of the observed change observed with temperature at 26 meV is due to phonons.

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