Superconducting and normal-state magnetic-susceptibility anisotropy in $YBa_2Cu_3O_7$

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Measurements of the superconducting and normal-state magnetic-susceptibility χ anisotropies of high-purity grain-aligned powder samples of $YBa_2Cu_3O_7$ are reported. Excellent alignment was achieved using a noncontaminating *in situ* technique. We find a strong temperature-dependent anisotropy $\Delta\chi \equiv \chi_c - \chi_{ab}$ that decreases from positive values for $T > T_c$ to negative values for $T < T_c$, consistent with reported torque-magnetometer measurements on single crystals. Estimates of the normal-state spin contributions to χ are obtained and compared with previous results.

INTRODUCTION

Study of the anisotropy in the normal-state thermodynamic and electronic transport properties can perhaps help to identify the relevant quasiparticle excitations in the metallic state of the high-T_c cuprates, because these anisotropies should depend in detail on the nature of these excitations. Although there have been many studies[1–4] of the normal-state anisotropy in the resistivity of single crystals, these have not led to an unambiguous identification of the nature of the quasiparticles. Thermodynamic measurements such as the magnetic susceptibility $\chi(T)$ would seem to be more directly amenable to theoretical analysis. Herein, we present measurements of the anisotropy in $\chi(T)$ for the superconducting as well as normal states of high-purity $YBa_2Cu_3O_7$, and compare the results with previous measurements on single crystals and powders. A brief outline of some of the present experimental results and a theoretical analysis of the anisotropic contribution to χ above T_c from superconducting fluctuation diamagnetism were given previously.[5]

EXPERIMENTAL DETAILS

A master batch of high-purity $YBa_2Cu_3O_7$ powder was prepared as described previously.[5] The grain size was measured using an optical microscope and was found to be quite uniform (diameter $\simeq 25$ μm). The grains were roughly cubic in shape, and appeared to be primarily well-formed single crystals. Magnetization data were obtained using a Quantum Design superconducting quantum interference device (SQUID) magnetometer. From magnetization versus field $M(H)$ isotherms, the ferromagnetic impurity contribution to the magnetizations was found to be small, equivalent to that of $\simeq 3$ ppm of iron metal impurities with respect to copper, and is corrected for in the χ data later.

As first pointed out by Farrell *et al.*,[6] grain alignment of $YBa_2Cu_3O_7$ powder can be achieved near room temperature by placing a free-flowing powder freshly mixed with epoxy in a strong magnetic field **H**; very good alignment with $\mathbf{c}\|\mathbf{H}$ is retained once the epoxy has cured. We first utilized this method with $H = 80$ kG at 300 K using Epotek 301 epoxy. Shown in Fig. 1 are $\chi(T)$ data for the epoxy alone ($H = 10$ kG), for the grain-aligned powder alone ($\mathbf{c}\|\mathbf{H}$, $H = 3$ kG, see below), and for 27 mg of the aligned powder in 9 mg of epoxy ($\mathbf{c}\|\mathbf{H}$, $H = 3$ kG). The Curie-Weiss contribution $[\chi = C/(T - \Theta)]$, evident in the latter data but not in the former two measurements, indicates that a chemical reaction between the epoxy and the sample occurred that generated paramagnetic species. Indeed, the negative curvature in $\chi(T)$ intrinsic to $YBa_2Cu_3O_7$ below ~ 200 K is completely masked by the paramagnetic impurity and/or defect contribution. We further observed for this aligned sample in epoxy that the screening susceptibility below T_c for $H = 50$ G was significantly degraded. The apparent deterioration of the superconductivty might be explained, e.g., by an influence of the epoxy on the coupling between grains, but the epoxy-induced Curie tail must originate from a chemical reaction between the sample and epoxy.

We therefore sought a better method of grain alignment. It is known that $\chi(T)$ for $\mathbf{H}\|\mathbf{c}$ (χ_c) is larger than that for $\mathbf{H}\perp\mathbf{c}$ near room temperature,[6,7] whereas $\chi_c < \chi_{ab}$ if $T < T_c$.[8,9] The free energy is minimized for $\mathbf{c}\|\mathbf{H}$ above T_c and for $\mathbf{c}\perp\mathbf{H}$ below T_c, and grain alignment will be in

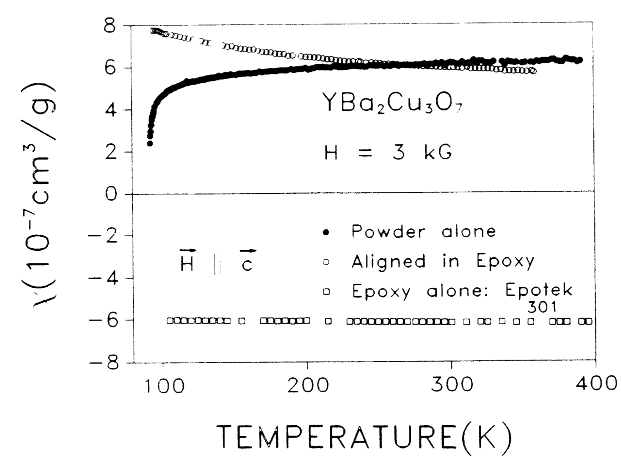

FIG. 1. Magnetic susceptibility χ vs temperature for grain-aligned $YBa_2Cu_3O_7$ powder ($\mathbf{H}\|\mathbf{c}$), for Epotek 301 epoxy, and for powder aligned in epoxy with $\mathbf{H}\|\mathbf{c}$.

these directions in the respective alignment temperature range if the grains are free to rotate. We therefore utilized an *in situ* method of grain alignment of our $YBa_2Cu_3O_7$ powder in the SQUID magnetometer. The free-flowing powder was placed in a quartz tube sample holder rigidly attached to the vertical sample rod. The rod was vibrated with a small 60 Hz buzzer attached to the top of the rod, outside the sample chamber of the magnetometer. Grain alignment was achieved in a field of 50 kG with $\mathbf{c}\|\mathbf{H}$ by holding the sample temperature at 300 K ($>T_c$), or with $\mathbf{c}\perp\mathbf{H}$ at 10 K ($<T_c$), and vibrating the sample rod overnight. The amplitude of vibration was then slowly (over minutes) reduced to zero and then the field reduced to zero. The subsequent magnetization measurements reported later were carried out in fields $H \le 5$ kG, fields low enough that grain realignment did not occur from 4 to 400 K for either field alignment direction. The obvious advantages of this alignment method over the above epoxy method are that magnetic impurities are not introduced into the sample upon grain alignment, and that the accuracy of the measured $\chi(T)$ anisotropy is not compromised by removing and/or handling the sample between measurements of the two field orientations.

We note that excessive grinding of our sample in air using an agate mortar and pestle apparently resulted in the generation of magnetic defects. This is illustrated in Fig. 2, which shows $\chi(T)$ data for powders grain aligned *in situ* with $\mathbf{c}\|\mathbf{H}$ before and after heavily grinding the powder. The grain size of the heavily ground sample was measured, as earlier, to be $\lesssim 1$ μm, much smaller than the aforementioned value of 25 μm measured prior to grinding. Although $\chi(T)$ for the heavily ground sample still shows negative curvature above T_c, the slope $d\chi/dT$ becomes negative above about 200 K; this indicates the presence of a Curie-Weiss contribution, presumably originating from magnetic defects generated during grinding.

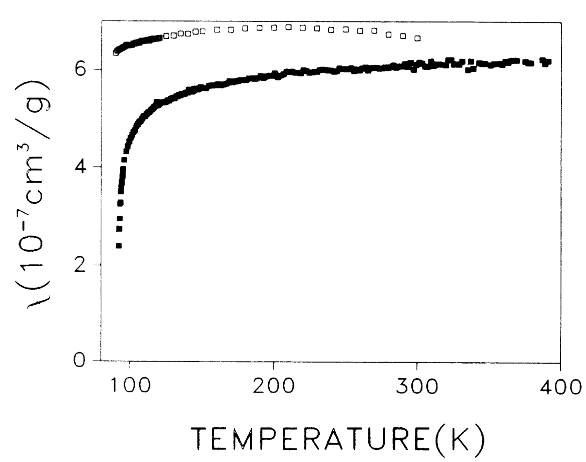

FIG. 2. Magnetic susceptibility χ vs temperature for grain-aligned $YBa_2Cu_3O_7$ with $\mathbf{H}\|\mathbf{c}$, before (closed squares) and after (open squares) heavily grinding the powder in air.

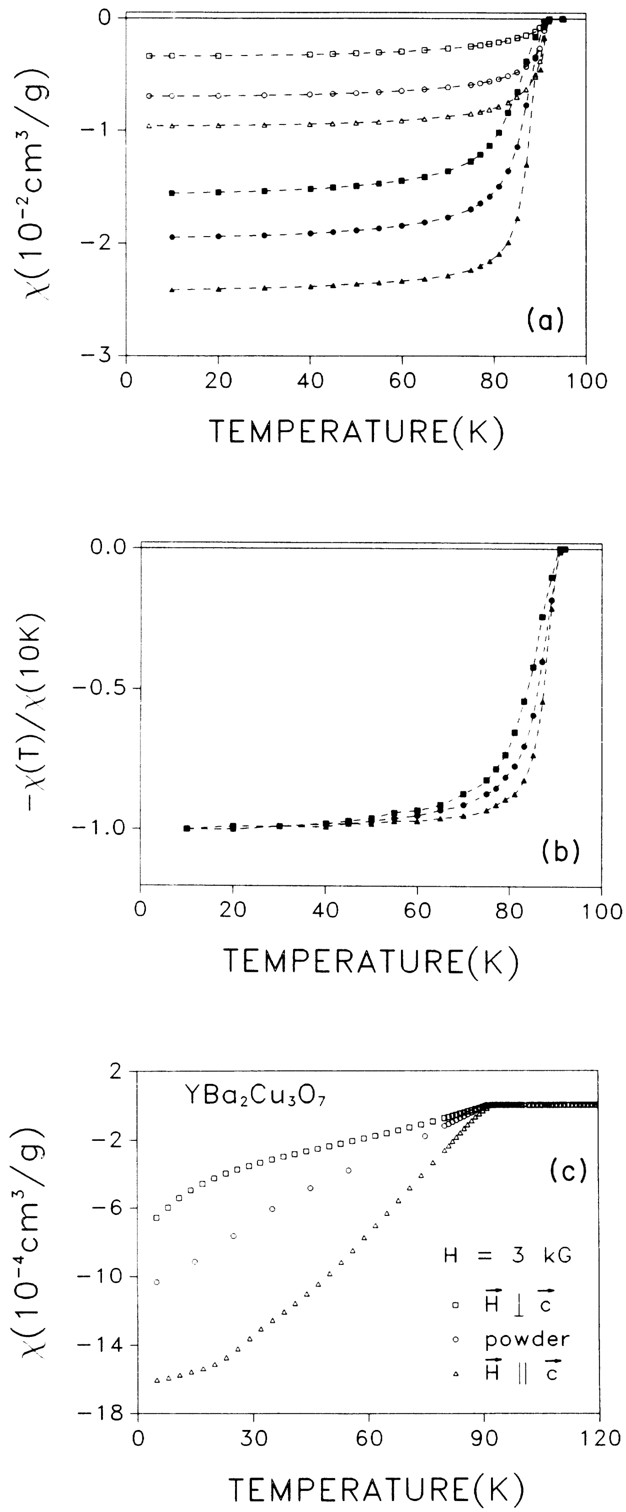

FIG. 3. (a) Screening susceptibility χ (closed symbols, zero-field cooled) and Meissner effect (open symbols, field cooled) vs temperature for nonaligned powder (circles) and grain-aligned samples of $YBa_2Cu_3O_7$ in a field of 50 G with $\mathbf{H}\|\mathbf{c}$ (triangles) and $\mathbf{H}\perp\mathbf{c}$ (squares). (b) Normalized magnetic susceptibility $-\chi(T)/\chi(10\text{ K})$ vs temperature for the screening measurements in (a). (c) Meissner effect χ measurements vs temperature in a field of 3 kG; the symbols are the same as in (a).

RESULTS

The screening diamagnetism (zero-field cooled) and Meissner effect (field cooled) were measured for our high-purity $YBa_2Cu_3O_7$ in a field of 50 G for both $\mathbf{H}\|\mathbf{c}$ and $\mathbf{H}\perp\mathbf{c}$, as well as for the randomly oriented powder. The results for $\chi(T)\equiv M(T)/H$ are shown in Fig. 3(a). Uncorrected for demagnetization factors, the randomly oriented powder shows 165% of $(-1/4\pi)$ screening susceptibility and a 42% Meissner effect. From Fig. 3(a), these measurements depend sensitively on the magnetic field orientation, as expected. The anisotropy in the screening susceptibility is comparable with that found for a single crystal;[10] however, because of variabilities associated with shape effects and possible vortex pinning at twins, one does not necessarily expect the Meissner and shielding results to be identical for aligned powders and single crystals. Figure 3(b) shows the screening susceptibilities for the three measurements in Fig. 3(a) normalized to the values at 10 K; for $\mathbf{H}\|\mathbf{c}$, the superconducting transition is seen to be narrower than those of the other two measurements. Meissner effect measurements in a field of 3 kG are shown in Fig. 3(c); the flux explusion is about an order of magnitude smaller than seen for $H=50$ G in Fig. 3(a), and the apparent transition width is of order T_c.

Magnetic-susceptibility $\chi(T)$ data were obtained above $T_c=91$ K in fields of 3 or 5 kG on nonaligned and aligned samples of mass 17–53 mg. Six different complete sets of data, as in Fig. 4, were obtained on four different samples from the same batch. The largest anisotropy in χ was observed for the sample with the smallest mass (17.6 mg). The $\chi(T)$ data for $H=3$ kG are shown in Fig. 4 for this $YBa_2Cu_3O_7$ sample before and after grain alignment. Another complete set of data for this sample closely reproduced the data in Fig. 4. The $\chi(T)$ data for each measurement increase monotonically with increasing temperature and exhibit negative curvature below ~ 200 K ($\sim 2T_c$). Above 240 K, the data increase approximately linearly with temperature, with slope $(1.40\pm0.08)\times10^{-10}$ cm^3/g K for both χ_c and χ_{ab}. The anisotropy $\Delta\chi\equiv\chi_c-\chi_{ab}$ is plotted in Fig. 5. Above T_c, $\Delta\chi>0$, whereas below T_c, $\Delta\chi<0$, as noted earlier. That $\Delta\chi$ passes through zero very near to T_c is consistent with the temperature-dependent anisotropy obtained from torque-magnetometer measurements on single crystals.[11] The χ_c, χ_{ab}, and $\Delta\chi$ values at 300 K are listed in Table I. The ratio $\chi_c/\chi_{ab}\simeq1.63$ at 300 K, and the temperature dependence of this ratio is shown in Fig. 6; for all temperatures except perhaps very close to T_c, $|\chi_c/\chi_{ab}|>1$.

DISCUSSION

We would first like to reemphasize[5] that the *observed* susceptibility data for $YBa_2Cu_3O_7$ in Fig. 4 exhibit no evidence for a Curie-Weiss contribution to $\chi(T)$. The data increase monotonically with temperature for both field orientations, exhibiting *negative* curvature below ~ 200 K. Between $\simeq 200$ and 400 K, our data increase approximately linearly with temperature, confirming the intrinsic behavior deduced previously based on a study of the variation of $\chi(T)$ with oxygen content in this system.[12] Other groups have also observed directly (without Curie-term correction) that $d\chi/dT>0$ above T_c.[13,14] However, most published data are dominated by the presence of magnetic defects and/or impurity phases (see also Figs. 1 and 2), yielding either nearly temperature-independent $\chi(T)$ behavior or $d\chi/dT<0$ above T_c. The negative curvature in our data from $T_c=91$ K to $\simeq 200$ K arises from a combination of superconducting fluctuation diamagnetism and a temperature-dependent normal-state background susceptibility.[5,12,13,15] Above $\simeq 200$ K, the superconducting fluctuation diamagnetism is negligible, and the temperature dependence of χ most likely arises from antiferromagnetic spin fluctuations.[12,16]

Of particular interest here is the anisotropy in $\chi(T)$ in $YBa_2Cu_3O_7$ above ~ 200 K. For comparison with the present measurements, we list in Table I values of χ_c, χ_{ab}, and $\Delta\chi$ for $YBa_2Cu_3O_7$ and for several other members of the high-T_c cuprate family obtained from other studies.[7,11,17–20] Because of the presence of vari-

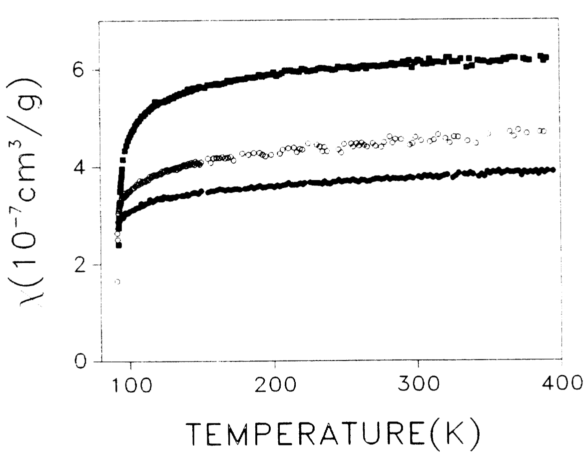

FIG. 4. Magnetic susceptibilities χ vs temperature in a field of 3 kG for $\mathbf{H}\|\mathbf{c}$ (squares), nonaligned powder (open circles), and for $\mathbf{H}\perp\mathbf{c}$ (closed circles).

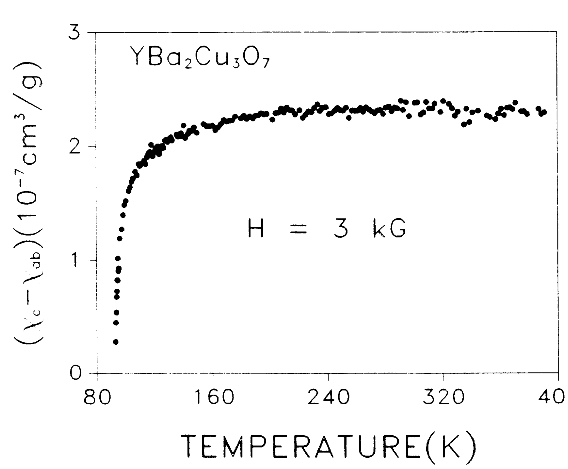

FIG. 5. Magnetic susceptibility anisotropy $\Delta\chi\equiv\chi_c-\chi_{ab}$ vs temperature for $YBa_2Cu_3O_7$.

TABLE I. Experimental magnetic susceptibility data for $\mathbf{H}\|\mathbf{c}$ (χ_c) and $\mathbf{H}\perp\mathbf{c}$ (χ_{ab}) at 300 K for $YBa_2Cu_3O_7$ and several other compounds. The superconducting transition temperature T_c, anisotropy $\Delta\chi \equiv \chi_c - \chi_{ab}$ and powder-averaged values $\langle\chi\rangle$ are also listed. The χ data of Refs. 7, 17, 18, and 20 are corrected for the contribution of paramagnetic impurities. The data for $Sr_2CuO_2Cl_2$ are for a temperature of 400 K, above the Néel temperature of $\simeq 310$ K (Ref. 20). An asterisk indicates units of 10^{-5} cm^3/mole.

Compound	T_c (K)	χ_c (*)	χ_{ab} (*)	$\Delta\chi$ (*)	$\langle\chi\rangle$ (*)	Reference
$YBa_2Cu_3O_7$	91	41.0	25.2	15.8	30.5	This work
$YBa_2Cu_3O_7$	87			13.8		11
$YBa_2Cu_3O_7$	87	41.3	28.5	12.8	32.8	7
$YBa_2Cu_3O_7$	92	33.9	25.0	8.9	28.0	17
$YBa_2Cu_3O_7$	90				29.3	18
$YBa_2Cu_3O_7$	92				27.7	18
La_2CuO_4		10.9	3.2	7.7	5.8	19
$La_{1.9}Sr_{0.1}CuO_4$	31	19.7	13.1	6.6	15.3	7
$Sr_2CuO_2Cl_2$		9.1	1.8	7.3	4.2	20

able amounts of paramagnetic and ferromagnetic impurities in various samples, which are sometimes not accounted for, perhaps the most reliable quantity to compare is $\Delta\chi$. Our value of $\Delta\chi$ for $YBa_2Cu_3O_7$ is slightly larger than those of Miljak *et al.*[11] and Fukuda *et al.*,[7] obtained for single crystals with a somewhat lower T_c, and is about 80% larger than recently reported[17] for a sample of $YBa_2Cu_3O_7$ grain-aligned powder in epoxy. From our large $\Delta\chi$ value, we conclude that the degree of grain alignment achieved in our experiments is essentially 100%; because our grain-aligned samples are free-flowing powders enclosed in quartz tubes, it was not possible to verify the degree of grain alignment using x-ray diffraction techniques. The reason that our value of $\Delta\chi$ is larger than observed[7,11] for the two single crystals may be that the substantial number of paramagnetic impurities present[7,11] in those crystals have anisotropic susceptibilities that partially cancel the anisotropy intrinsic to $YBa_2Cu_3O_7$. A comparison of $\Delta\chi$ for a ceramic compact with our value and those in Ref. 11 has been useful in estimating the degree of grain alignment in the compact.[21]

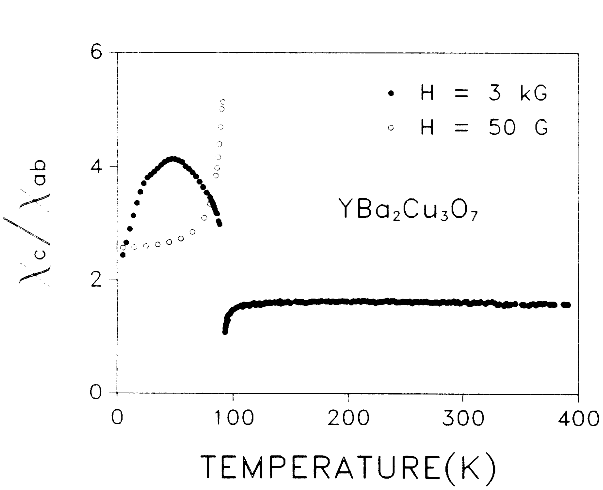

FIG. 6. Ratio of the susceptibility with the field H parallel and perpendicular to the c axis χ_c/χ_{ab} vs temperature for $YBa_2Cu_3O_7$.

It is noteworthy that the $\Delta\chi$ values for nonmetallic La_2CuO_4 and $Sr_2CuO_2Cl_2$ and for metallic $La_{1.88}Sr_{0.12}CuO_4$ in Table I are the same to within $\pm 7\%$. This similarity suggests that the local electronic structure around the Cu ions in the CuO_2 planes is similar in these compounds.

To analyze our data in Table I, we separate χ for each field direction into orbital and spin contributions:

$$\chi(T) = \chi^{\mathrm{orb}} + \chi^{\mathrm{spin}}(T) , \tag{1}$$

where χ^{orb} is assumed independent of temperature, whereas χ^{spin} may depend on temperature. In the absence of superconducting fluctuation diamagnetism (i.e., above $\simeq 200$ K), we assume

$$\chi^{\mathrm{orb}} = \chi^{\mathrm{dia}} + \chi^{\mathrm{VV}} , \tag{2}$$

which consists of the isotropic core diamagnetism χ^{dia} and the paramagnetic, and in general anisotropic, Van Vleck contribution χ^{VV} (we absorb possible contributions from Landau diamagnetism into χ^{spin}). From Eqs. (1) and (2),

$$\Delta\chi \equiv \chi_c - \chi_{ab} = \Delta\chi^{\mathrm{VV}} + \Delta\chi^{\mathrm{spin}} . \tag{3}$$

Estimates of χ^{spin} and χ^{VV} have been made previously for $YBa_2Cu_3O_7$. From the observed variation in the heat-capacity jump at T_c with derived temperature-independent susceptibility, Junod *et al.*[18] derived the values in Table II. Analysis of the shifts from nuclear resonance experiments yielded other estimates, shown in Table II.[17,22,23] Also shown in Table II are the predictions from band theory for χ^{dia} and χ^{VV} of hypothetical Sc_2CuO_4;[24] the χ^{VV} values are smaller than inferred for the Cu(2) ions in the CuO_2 planes of $YBa_2Cu_3O_7$.

Takiegawa *et al.*[17] derived their values of χ^{spin} using their anisotropic χ data in Table I. Since we infer that the intrinsic anisotropy is much larger than reported in Ref. 17, we have recomputed χ^{spin} using Eqs. (1) and (2), their values for χ^{dia} and χ^{VV}, and our χ anisotropy data, and the results are shown in Table II. The spin susceptibility is larger by about 10% for $\mathbf{H}\|\mathbf{c}$ than for $\mathbf{H}\perp\mathbf{c}$, rath-

TABLE II. Derived contributions to the susceptibility, all in units of 10^{-5} cm^3/mole. Cu(2) and Cu(1) refer, respectively, to the Cu in the planes and chains of $YBa_2Cu_3O_7$.

Entity	χ^{dia}	χ_c^{VV}	χ_b^{VV}	χ_a^{VV}	$\Delta\chi^{VV}$	$\langle\chi^{VV}\rangle$	χ^{spin}	Reference
$YBa_2Cu_3O_7$	−17.5					14.5	32.5	11
$YBa_2Cu_3O_7$	−17.5					≥22.9	≤21.6	22
Cu(2)		12.7	2.9	2.9	9.8	6.2	18.8	23
Cu(2)		9.6(6)	1.7(2)	1.7(2)	7.9(7)	4.3(3)		17
Cu(1)		2.2(6)	3.0(2)	8.4(2)	−3.5(7)	4.5(3)		17
$YBa_2Cu_3O_7$	−17.5	21.3(10)	6.4(4)	11.8(4)	12.2(12)	13.2(12)	30.1, **H**‖c 33.4, **H**⊥c	17
$YBa_2Cu_3O_7$	−17.5	21.3(10)	6.4(4)	11.8(4)	12.2(12)	13.2(12)	37.2, **H**‖c 33.6, **H**⊥c	This work
Sc_2CuO_4	−11	4.0	1.5	1.5	2.5	2.3		24

er than the reverse,[17] consistent with the sign of $\Delta\chi^{spin}$ in $Sr_2CuO_2Cl_2$;[20] the latter anisotropy probably arises from anisotropy in the spectroscopic splitting factor g of the spin-$\frac{1}{2}$ Cu^{2+} ions.[20] We note that all the derived values of χ^{spin} for $YBa_2Cu_3O_7$ in Table II would shift upwards by 1.8×10^{-5} cm^3/mole if the value of χ^{dia} computed from Ref. 25 (-19.3×10^{-5} cm^3/mole) were substituted for the value in Table II.

CONCLUDING REMARKS

We have shown that the intrinsic magnetic susceptibility χ of $YBa_2Cu_3O_7$ is highly anisotropic and is strongly temperature dependent below 200 K, exhibiting negative curvature. For **H**‖c as well as **H**⊥c, χ increases monotonically with temperature from T_c up to at least 400 K, in agreement with the analysis of Ref. 12. The strong negative curvature in $\chi(T)$ and its anisotropy below ~200 K were found in Ref. 5 to arise from a combination of superconducting fluctuation diamagnetism and a temperature-dependent normal-state susceptibility. The powder-averaged value of χ at 300 K is close to the corrected values inferred for the best samples in the systematic study of Junod *et al.*[18] The spin susceptibility χ^{spin} was derived using the orbital susceptibility values for Cu^{2+} ions inferred from the nuclear resonance measurements of Takigawa *et al.*,[17] and found to be nearly isotropic, in agreement with those authors. Thus, most of the large anisotropy in χ above ~200 K is of orbital origin, arising from the Cu d orbitals.[17] The anisotropy $\Delta\chi^{VV}$ in the orbital χ found[17] for Cu(2) in the CuO_2 layers of $YBa_2Cu_3O_7$ (Table II) is about the same as observed for nonmetallic La_2CuO_4 (Ref. 19) and $Sr_2CuO_2Cl_2$ (Ref. 20), and for superconducting metallic $La_{1.9}Sr_{0.1}CuO_4$ (Ref. 7) (Table I), where the latter three compounds contain CuO_2 layers but no Cu-O chain layers. This similarity suggests that the local electronic environments around the Cu ions in the CuO_2 layers of all four compounds are similar, despite the fact that two of the compounds exhibit a metallic character, whereas the other two do not. The powder-averaged χ^{spin} (at 300 K) and χ^{VV} for $YBa_2Cu_3O_7$ are remarkably close to the values inferred from the variation in the heat-capacity jump at T_c with corrected χ by Junod *et al.*[11] (see Table II). A detailed understanding of this agreement must await further theoretical advances regarding the superconducting and normal states.

Note added in proof. The anisotropy in $\chi(T)$ for grain aligned $YBa_2Cu_3O_y$ ($y\approx6.1-7.0$) has also recently been studied by Yamaguchi *et al.*[26]

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