RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 42, NUMBER 13 1 NOVEMBER 1990

Magnetic-susceptibility anisotropy of single-crystal $Bi_2Sr_2CaCu_2O_8$

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(Received 13 July 1990)

Magnetic susceptibility $\chi(T)$ data from 84 to 300 K are reported for a $Bi_2Sr_2CaCu_2O_8$ single crystal. The $\chi(T)$ data with $\mathbf{H}\|\mathbf{c}$ exhibit negative curvature up to $\simeq$150 K, attributed to superconducting-fluctuation diamagnetism, whereas those with $\mathbf{H}\perp\mathbf{c}$ are independent of temperature above 90 K. From a theoretical fit to the data with $\mathbf{H}\|\mathbf{c}$, the Ginzburg-Landau coherence length $\xi_{ab}(0)$ is found to be 10.9 Å. The room-temperature orbital and spin susceptibilities and their anisotropies are estimated and compared with those of $YBa_2Cu_3O_7$.

The magnetic susceptibility $\chi(T)$ has been found to be strongly anisotropic in La_2CuO_4, $Sr_2CuO_2Cl_2$, $La_{2-x}Sr_xCuO_4$, and $YBa_2Cu_3O_{7-\delta}$, with the normal-state susceptibility with $\mathbf{H}\|\mathbf{c}$ ($\chi_\|$) greater than for $\mathbf{H}\perp\mathbf{c}$ ($\chi_\perp$).[1] For most of the compounds, both $\chi_\|$ and $\chi_\perp$ increase monotonically with increasing temperature at the higher temperatures; for the first two of these, which are antiferromagnetic insulators, this behavior is as expected for the two-dimensional Heisenberg antiferromagnet on a square lattice for temperatures $T \lesssim J$, where the exchange coupling between nearest-neighbor Cu^{2+} spins $\frac{1}{2}$ is $\mathbf{J}\mathbf{S}_i\cdot\mathbf{S}_j$.[2] For the last two superconducting compounds, the behavior arises from a combination of antiferromagnetic spin correlations and superconducting-fluctuation diamagnetism (SFD).[3,4] Amazingly, the molar anisotropy $\Delta\chi \equiv \chi_\| - \chi_\perp$ per CuO_2 plane at high temperatures ($\simeq$300–400 K) is very similar in each of the four systems [$\Delta\chi \simeq (9\pm2)\times10^{-5}$ cm^3/mol CuO_2],[1] despite the fact that the first two are insulators and the last two exhibit metallic and superconducting properties. This suggests that the electronic environments around the Cu atoms in the CuO_2 planes are similar in each system.

Herein, we report $\chi_\|(T)$ and $\chi_\perp(T)$ from $T_c \simeq 84$ K to 300 K of a 2.33-mg single crystal of $Bi_2Sr_2CaCu_2O_8$. Our values are consistent with powder data,[3,5] and $\chi_\|(T)$ shows negative curvature below $\simeq$150 K, similar to that seen for the powders and attributed to SFD. From analysis of these data, estimates of the zero-temperature Ginzburg-Landau coherence length parallel to the CuO_2 planes, $\xi_{ab}(0)$, and the room-temperature orbital and spin susceptibilities and their anisotropies are obtained and compared with corresponding values for $YBa_2Cu_3O_7$.

The $Bi_2Sr_2CaCu_2O_8$ crystal was grown using the self-flux method by heating the stoichiometric mixture of oxides to 950 °C and slowly cooling. The $\chi(T)$ data were obtained using a Quantum Design superconducting quantum interference device (SQUID) magnetometer in a field of 15 kG. Meissner-effect data in a field of 50 G with $\mathbf{H}\|\mathbf{c}$ showed a transition onset at 85.0 K, a midpoint of 84.0 K and a 10%–50% width of 0.8 K, with a Meissner fraction at 5 K of 91%. Zero-field-cooled data in the same field showed a volume susceptibility of 240% of $-1/4\pi$; both values are uncorrected for demagnetization factors. With $\mathbf{H}\perp\mathbf{c}$, the corresponding fractions were 4.2% and 50%.

The magnetic susceptibilities $\chi_\|(T)$ and $\chi_\perp(T)$ for temperatures $T \gtrsim T_c$ are shown in Fig. 1(a). $\chi_\|(T)$ exhibits negative curvature from T_c up to $\simeq$150 K, then is nearly independent of temperature up to 300 K, increasing linearly at the small rate 1.5×10^{-10} cm^3/gK. On the other hand, to within the experimental precision, $\chi_\perp$ is independent of temperature from $\simeq$90 to 300 K. This is expected. The superpair effective-mass ratio for this compound is $\sim 3\times10^3$ (Ref. 6), so the SFD for $\mathbf{H}\perp\mathbf{c}$ should be less than that for $\mathbf{H}\|\mathbf{c}$ by the square root of this fac-

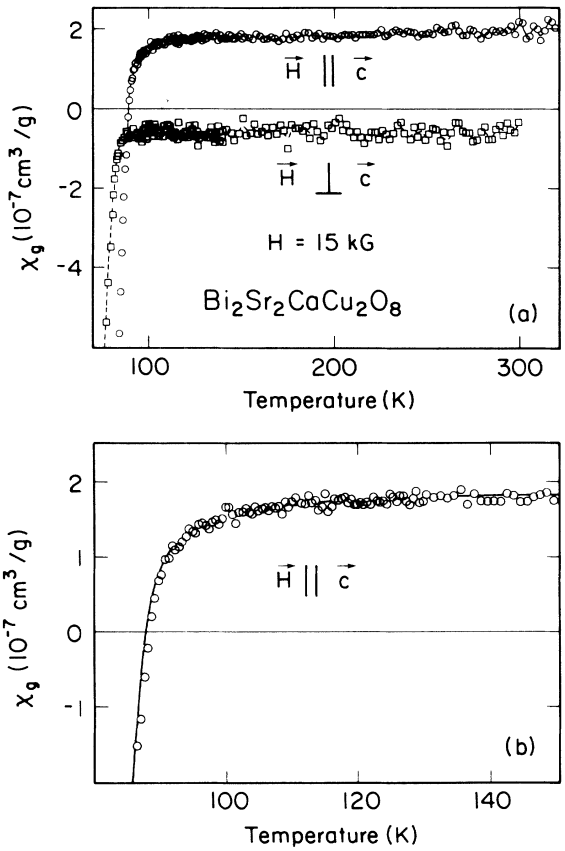

FIG. 1. (a) Magnetic susceptibility χ_g vs temperature for a 2.33-mg single crystal of $Bi_2Sr_2CaCu_2O_8$ for $\mathbf{H}\|\mathbf{c}(\chi_\|)$ and $\mathbf{H}\perp\mathbf{c}(\chi_\perp)$. (b) Expanded plot of the data for $\mathbf{H}\|\mathbf{c}$ in (a) below 150 K. The solid curve is a fit of superconducting fluctuation diamagnetism theory (Refs. 3 and 7–9) to the data above 90 K (see text).

tor,[3] i.e., should be unobservable in our measurements except for very close to T_c. From Fig. 1(a), $\Delta\chi$ decreases with decreasing temperature and becomes negative below 87.4 K, which is about 3 K above T_c as determined above. This crossover occurs significantly above the bulk T_c because the SFD is increasing much faster with decreasing temperature for **H**$\|$**c** than for **H**$\perp$**c**, as just noted, and because the sensitivity of the magnetometer is much greater for $H=1.5$ T than for $H=50$ G. The values of $\chi_\|$, $\chi_\perp$, $\Delta\chi$, and $\langle\chi\rangle$ at 300 K are listed in Table I. The powder data for $Bi_{2-x}Pb_xSr_2CaCu_2O_8$ at 300 K in Refs. 3 and 5 lie between our $\chi_\|$ and $\chi_\perp$ values, but are above the powder average in Table I; this suggests preferred orientation of the powders with **c**$\|$**H**.

An expanded plot of the $\chi_\|(T)$ data from T_c to 150 K is shown in Fig. 1(b). We fitted the data above 90 K to the expression $\chi(T)=\chi_0+\chi_{fl}(T)$ using the Lawrence-Doniach theory[7] as modified by Klemm[3,8] for the SFD, $\chi_{fl}(T)$, in the two-dimensional low-field regime with **H**$\|$**c**:

$$\chi_{fl}(T) = -[g_{eff}\pi k_B \xi_{ab}^2(0)T/3\phi_0^2 s][T_c/(T-T_c)] , \qquad (1)$$

where $T_c \equiv T_c(H)$, $g_{eff}=2$ is the number of independently fluctuating CuO_2 layers per CuO_2-layer repeat distance $s=15.4$ Å, ϕ_0 is the flux quantum $hc/2e$, and χ_0 is the background susceptibility. We believe that our data are in, or close to, the low-field regime, since the applied field $H=1.5$ T $\ll H_{c2}(0)\sim 100$ T.[3,8,9] The fitting parameters obtained were $T_c=(84.6\pm 1)$ K, $\chi_0=(1.97\mp 0.02)\times 10^{-7}$ cm^3/g, and $\xi_{ab}(0)=(10.9\mp 0.9)$ Å. The fit is shown as the solid curve in Fig. 1(b). From the value of $\xi_{ab}(0)$, one obtains $H_{c2}(0)=\phi_0/2\pi\xi_{ab}^2(0)\sim 280$ T, consistent with $H \ll H_{c2}(0)$ and with the small inferred value of $T_c(0)-T_c(H=1.5\text{ T})$. The inferred value of $\xi_{ab}(0)$ would increase and $H_{c2}(0)$ would decrease if the data in Fig. 1(b) were corrected for a Curie-like term due to undetected magnetic impurities and/or isolated Cu^{2+} defects; the inferred $T_c(H)$ would also be affected. Thus, our $\xi_{ab}(0)$ and $H_{c2}(0)$ values should be considered as lower and upper limits, respectively. $\xi_{ab}(0)$ is about the same as that for $YBa_2Cu_3O_7$ (13.6 Å) (Ref. 3) and $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_{10}$ (11.8–18.0 Å),[5] but is about one-half the value previously inferred for $Bi_{2-x}Pb_xSr_2CaCu_2O_8$ (20.4 Å),[5] where all of these $\xi_{ab}(0)$ values were derived from fits to $\chi_{fl}(T)$ data. The discrepancy between our $\xi_{ab}(0)$ value and that in Ref. 5 might be partially explained if the grains in the powder sample studied there were preferentially aligned to some extent with **c**$\|$**H**, as

TABLE I. Magnetic susceptibility data at 300 K for single-crystal $Bi_2Sr_2CaCu_2O_8$ (this work) and grain-aligned high-purity polycrystalline $YBa_2Cu_3O_7$ (Ref. 1). $\chi_\|$ and $\chi_\perp$ are, respectively, the susceptibility with **H**$\|$**c** and **H**$\perp$**c** at 300 K, $\Delta\chi\equiv\chi_\|-\chi_\perp$, $\langle\chi\rangle$ is the powder-averaged value, χ^{core} the atomic core diamagnetism, K_α^L the orbital Knight shifts for ^{63}Cu in $Bi_2Sr_2CaCu_2O_8$ derived here, and in $YBa_2Cu_3O_7$ from Ref. 10, χ_α^{VV} the derived Van Vleck susceptibilities of the Cu^{2+} ions, χ_α^{spin} the derived spin susceptibilities, and $g_\|/g_\perp \equiv (\chi_\|^{spin}/\chi_\perp^{spin})^{1/2}$ the ratio of the spectroscopic splitting factors. The g_α values ($\alpha=a,b,c$) were computed independently using the corresponding χ_α^{VV} values. All susceptibilities are in units of 10^{-5} cm^3/mol. In $YBa_2Cu_3O_7$, Cu(1) is in the Cu-O chains and Cu(2) is in the CuO_2 planes. Note that the χ^{spin} values for $YBa_2Cu_3O_7$ include the contributions from Cu in both the CuO_2 planes and Cu-O chains, whereas $Bi_2Sr_2CaCu_2O_8$ contains no Cu-O chains.

Entity	$Bi_2Sr_2CaCu_2O_8$	$YBa_2Cu_3O_7$
$\chi_\|$	17.1(10)	41.0
$\chi_\perp$	−5.4(13)	25.2
$\Delta\chi$	22.5(16)	15.8
$\langle\chi\rangle$	2.1(16)	30.5
χ^{core}	−20.6	−17.5
K_c^L(%)	1.79	1.28(1)[Cu(2)],0.25(1)[Cu(1)]
K_b^L(%)	0.45	0.28(2)[Cu(2)],0.27(4)[Cu(1)]
K_a^L(%)	0.45	0.28(2)[Cu(2)],1.08(4)[Cu(1)]
χ_a^{VV}[Cu(1)]	· · ·	8.1
χ_b^{VV}[Cu(1)]	· · ·	2.0
χ_c^{VV}[Cu(1)]	· · ·	1.9
$\chi_\|^{VV}$[Cu(2)]	13.4	9.6
$\chi_\perp^{VV}$[Cu(2)]	3.3	2.1
$\chi_\|^{VV}$	26.7	21.1
$\chi_\perp^{VV}$	6.7	9.2
$\chi_\|^{spin}$	11.0	37.4
$\chi_\perp^{spin}$	8.5	33.5
$g_\|/g_\perp$	1.14	1.06
g_a	2.09	2.06[Cu(2)],2.22[Cu(1)]
g_b	2.09	2.06[Cu(2)],2.06[Cu(1)]
g_c	2.36	2.26[Cu(2)],2.06[Cu(1)]
$\bar{g}_c$	2.36	2.20
$\bar{g}_{a,b}$	2.09	2.10
$\bar{g}_c/\bar{g}_{a,b}$	1.13	1.05

inferred above, whereas random alignment was assumed.[5] Also, the superconducting transition width of the crystal is much smaller than in the powders.[5] Therefore, we believe the value for the single crystal studied here may be the more reliable.

Above $\simeq 200$ K where the superconducting fluctuation diamagnetism is negligible, a principal value $\chi_\alpha(T)$ is written as the sum of an orbital term and a spin term:

$$\chi_\alpha = \chi_\alpha^{\rm orb} + \chi_\alpha^{\rm spin} . \tag{2}$$

Both terms are anisotropic in general. In a localized moment picture for the Cu^{2+} spins $\frac{1}{2}$,[10] the orbital terms are the sum of the isotropic core diamagnetism $\chi^{\rm core}$ and the anisotropic Van Vleck paramagnetism $\chi_\alpha^{\rm VV}$ of the Cu^{2+} cations. The $\chi_\alpha^{\rm VV}$ per mole of Cu^{2+} is given by

$$\chi_\alpha^{\rm VV} = 2N_A\mu_B^2\Lambda_\alpha , \tag{3}$$

where

$$\Lambda_\alpha = \sum_n |\langle n|L_\alpha|0\rangle|^2/\Delta E_n ,$$

N_A is Avogadro's number, μ_B is the Bohr magneton, $\Delta E_n \equiv E_n - E_0$, L_α is the angular momentum operator in the αth principal direction, $|0\rangle \equiv |x^2-y^2\rangle$ is the crystal-field ground hole state of the Cu^{2+} ion with energy E_0, and the excited states $|n\rangle$ are $|xy\rangle$, $|xz\rangle$, and $|yz\rangle$ (the matrix elements to the state $|z^2-r^2\rangle$ all vanish). The Λ_α values are

$$\Lambda_z = 4/\Delta E_{xy},\ \Lambda_x = 1/\Delta E_{yz},\ \text{and}\ \Lambda_y = 1/\Delta E_{xz} \tag{4}$$

if the uniaxial direction is z. The spectroscopic splitting factors g_α of the Cu^{2+} ions are related to the (same) Λ_α values via

$$g_\alpha = 2(1-\lambda\Lambda_\alpha) , \tag{5}$$

where $\lambda = -710\ {\rm cm}^{-1} = -88.2$ meV is the spin-orbit coupling parameter for Cu^{2+}.[10] The spin susceptibilities $\chi_\alpha^{\rm spin}(T)$ per mole of formula units are written as

$$\chi_\alpha^{\rm spin}(T) = n_f N_A g_\alpha^2 \mu_B^2 F/J , \tag{6}$$

where here n_f is the number of CuO_2-plane units per formula unit, F is a dimensionless function of temperature which is the same for different α, and J is a characteristic energy. In insulating local moment antiferromagnets like La_2CuO_4, $Sr_2CuO_2Cl_2$, or $YBa_2Cu_3O_6$, the parameter $J \sim 1500$ K is the antiferromagnetic intralayer exchange coupling constant, where the nearest-neighbor exchange energy is $\mathbf{J}\mathbf{S}_i\cdot\mathbf{S}_j$, and $F = F(T/J)$. For $Bi_2Sr_2CaCu_2O_8$, Cu nuclear resonance shift data are not yet available to aid in computing the $\chi_\alpha^{\rm VV}$ and/or $\chi_\alpha^{\rm spin}$ terms, and thereby evaluate the ΔE_n values. We therefore assume that the relevant ΔE_n values are the same $\equiv\Delta$. The susceptibility is uniaxial. Gathering together the above terms into Eq. (2) yields

$$\chi_\parallel = \chi^{\rm core} + 8n_f N_A\mu_B^2/\Delta + 4n_f N_A\mu_B^2(1-4\lambda/\Delta)^2 F/J$$

and (7)

$$\chi_\perp = \chi^{\rm core} + 2n_f N_A\mu_B^2/\Delta + 4n_f N_A\mu_B^2(1-\lambda/\Delta)^2 F/J ,$$

where $\chi_\parallel$ refers to the uniaxial (**c**) direction. Multiplying Eqs. (7) by λ and defining dimensionless susceptibilities

$$\chi_\parallel' = \lambda(\chi_\parallel - \chi^{\rm core})/4n_f N_A\mu_B^2$$

and (8)

$$\chi_\perp' = \lambda(\chi_\perp - \chi^{\rm core})/4n_f N_A\mu_B^2 ,$$

Eqs. (7) become

$$\chi_\parallel' = 2r + (1-4r)^2 F\lambda/J$$

and (9)

$$\chi_\perp' = r/2 + (1-r)^2 F\lambda/J ,$$

where $r \equiv \lambda/\Delta$. Eliminating $F\lambda/J$ from Eqs. (9) yields a cubic equation for r:

$$6r^3 + a_2r^2 + a_1r + a_0 = 0 , \tag{10}$$

where

$$a_0 = \chi_\parallel' - \chi_\perp' ,$$

$$a_1 = 8\chi_\perp' - 2\chi_\parallel' - \tfrac{3}{2} ,$$

and

$$a_2 = \chi_\parallel' - 16\chi_\perp' .$$

Solving the first of Eqs. (9) for $F\lambda/J$ yields

$$F\lambda/J = (\chi_\parallel' - 2r)/(1-4r)^2 . \tag{11}$$

We take the $\chi^{\rm core}$ values as -25, -15, -8, -12, and -12×10^{-6} cm^3 per mole of Bi, Sr, Ca, Cu, and O, respectively,[11] giving the molar $\chi^{\rm core}$ value for $Bi_2Sr_2CaCu_2O_8$ ($n_f = 2$) shown in Table I. Using the above λ and the χ_α data in Table I, Eq. (10) predicts $r = -0.0455$ and $\Delta = 1.94$ eV. The $\chi_\alpha^{\rm VV}$ [Eq. (3)] and g_α [Eq. (5)] ($\alpha = \parallel, \perp$) values for $Bi_2Sr_2CaCu_2O_8$ were computed using these λ and Δ values and are listed in Table I. From Eq. (11) and the value of r, one obtains $F\lambda/J = -0.0266$, and $F/J = 0.301$ eV^{-1}. The $\chi_\alpha^{\rm spin}$ values computed from Eq. (6) are listed in Table I. Also listed are the orbital ^{63}Cu NMR shifts predicted using the $\chi_\alpha^{\rm VV}$ values in Table I and Eq. (12) below. In Table I, $g_\parallel/g_\perp \equiv (\chi_\parallel^{\rm spin}/\chi_\perp^{\rm spin})^{1/2} = 1.14$ [cf. Eq. (6)]. This ratio is computed independently from the g_α values ($\alpha = a,b,c$) to be 1.13, nearly the same. This agreement strongly supports the local moment picture for Cu^{2+} used here and (e.g., Ref. 10) elsewhere.

A similar analysis for $YBa_2Cu_3O_7$ has been carried out using NMR shift data.[10] The anisotropic $\chi_\alpha(300\ {\rm K})$ data from Ref. 1 are listed in Table I. The $\chi_\alpha^{\rm VV}$ values per mole of Cu can be estimated from the ^{63}Cu-NMR orbital shifts K_α^L of the Cu(1) (in the Cu-O chains) and Cu(2) (in the CuO_2 planes) ions in $YBa_2Cu_3O_7$ at 4.2 K according to[10]

$$K_\alpha^L = 2N_A\langle 1/r^3\rangle\chi_\alpha^{\rm VV} , \tag{12}$$

as shown in Table I, where $\langle 1/r^3\rangle = 6.0/(5.3\times10^{-9}\ {\rm cm})^3$ (Ref. 10). Using Eqs. (3) an (4) and the $\chi_\alpha^{\rm VV}$ values for $YBa_2Cu_3O_7$ in Table I, the ΔE_n values are found to be $\Delta E_{xy}(2) = 2.7$ eV and $\Delta E_{xz}(2) = \Delta E_{yz}(2) = 3.1$ eV for Cu(2), where $\mathbf{z} = \mathbf{c}$, $\mathbf{x} = \mathbf{a}$, and $\mathbf{y} = \mathbf{b}$.[10] For Cu(1), the uniaxial (**z**) crystal-field axis is along **a**; noting this, we have $\Delta E_{xz} \simeq \Delta E_{yz} = 3.2$ eV, and $\Delta E_{xy} = 3.2$ eV. From Eq.

(5), the predicted g_a values for Cu(1) and Cu(2) in $YBa_2Cu_3O_7$ are found and listed in Table I, referred to the **a**, **b**, and **c** crystal axes. From the measured values of χ_a and the deduced values of χ_a^{orb}, the values of χ_a^{spin} were computed from Eq. (2) and are listed in Table I.[12] The listed value of $g_\parallel/g_\perp \equiv (\chi_\parallel^{spin}/\chi_\perp^{spin})^{1/2} = 1.06$. The average values $\bar{g}_a$ derived from the listed g_a values ($a = a,b,c$) are also listed, where it is seen that $\bar{g}_c/\bar{g}_{a,b} = 1.05$, close to the value of 1.06 derived independently above. Thus, the anisotropies in both χ^{spin} and χ^{VV} in both $Bi_2Sr_2CaCu_2O_8$ and $YBa_2Cu_3O_7$ are quantitatively and self-consistently accounted for in the localized picture. Note that both the orbital and spin susceptibility anisotropies of $YBa_2Cu_3O_7$ are reduced from the values for the CuO_2 layers alone, because the respective anisotropies of the Cu in the Cu-O chains partially cancel those due to Cu in the CuO_2 layers. We remark that a χ^{spin} anisotropy similar to those in Table I was inferred for the tetragonal insulating antiferromagnet ($T_N \simeq 300$ K) $Sr_2CuO_2Cl_2$ above T_N ($g_c = 2.46$, $g_{ab} = 2.01$),[13] which has the same CuO_2 layers as in the other layered cuprates.

From Table I, the magnitudes of χ_a^{spin} are smaller in $Bi_2Sr_2CaCu_2O_8$ than in $YBa_2Cu_3O_7$ by factors of 3–4, even though the respective T_c's are within $\simeq 8\%$ of each other. Even if the χ_a^{VV} values in the former compound are taken to be identical to those of Cu(2) in the latter, the χ_a^{spin} values are still found to be smaller by factors of 2–3. What this means is unclear. At first sight, the similarity of T_c and the differences in χ_a^{spin} between the two compounds suggest that the mechanism for superconductivity in these materials involves factors other than, or in addition to, the magnetic character of the Cu^{2+} ions.

In conclusion, we find a substantial temperature-dependent anisotropy in the susceptibility of $Bi_2Sr_2CaCu_2O_8$ between $T_c \simeq 84$ K and $\simeq 150$ K with $\chi_\perp < \chi_\parallel$ for $T \gtrsim T_c$, as is the case for all cuprate superconductors (not containing magnetic ions other than Cu^{2+}) studied to date. The temperature dependence of the anisotropy arises primarily from that of the anisotropy in the superconducting fluctuation diamagnetism; from a fit with theory, we find $\xi_{ab}(0) = 10.9$ Å. The room-temperature value for $\chi_\perp$ is in agreement with the recent data of Ref. 14, but our $\chi_\parallel$ is much larger. The reason for this discrepancy is not known, but we note that the data of Ref. 14 are also inconsistent with powder data.[3,5] Assuming a local moment picture for the Cu^{2+} ions, where the system has a single spin degree of freedom, the anisotropy $\Delta\chi \equiv \chi_\parallel - \chi_\perp$ at 300 K is found to arise from anisotropy in both the Van Vleck susceptibility χ^{VV} of the Cu^{2+} ions and from anisotropy in the spin susceptibility χ^{spin}. About 90% of $\Delta\chi$ arises from the former anisotropy and the remainder from an anisotropic χ^{spin} originating from an anisotropic g factor of the Cu^{2+} ions. The magnitudes of χ_a^{spin} in $Bi_2Sr_2CaCu_2O_8$ are three to four times smaller than in $YBa_2Cu_3O_7$, even though the respective T_c's are quite comparable. The origin of these differences in χ_a^{spin} and their bearing on the T_c's are interesting issues for future theoretical and experimental clarification.

Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-Eng-82. This work was supported by the Director for Energy Research, Office of Basic Energy Sciences.

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