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Abstract
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Disciplines
Physics

Comments
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Direct Observation of the Quantum Energy Gap in $S{=}\frac{1}{2}$ Tetragonal Cuprate Antiferromagnets

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Using an electron spin resonance spectrometer covering a wide range of frequency and magnetic field, we have measured the low energy excitations of the $S{=}\frac{1}{2}$ tetragonal antiferromagnets, $Sr_2CuO_2Cl_2$ and $Sr_2Cu_3O_4Cl_2$. Our observation of in-plane energy gaps of order 0.1 meV at zero external magnetic field are consistent with a spin wave calculation, which includes several kinds of quantum fluctuations that remove frustration. Results agree with other experiments and with exchange anisotropy parameters determined from a five band Hubbard model.

76.50.+g, 75.10.Jm, 75.50.Ee

The fact that many systems containing copper oxide planes become high-T_c superconductors when suitably doped [1] has led to a continuing effort to understand in detail the magnetic properties of the undoped parent systems. Most of these systems contain weakly coupled CuO_2 planes, and the $S = \frac{1}{2}$ spins $\mathbf{S}_i$ on the Cu ions are well described as an antiferromagnet governed by the Hamiltonian $\mathcal{H} = \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{J}_{ij} \cdot \mathbf{S}_j$, where $\mathbf{J}_{ij}$ is the tensor for exchange interactions between ions i and j, and $\langle ij \rangle$ restricts the sum to pairs of nearest neighboring Cu spins. Above the Néel temperature $T_{\rm N}$, most experiments can be fully explained by specializing to only intraplanar isotropic coupling, so that $\mathbf{J}_{ij} = J\mathcal{I}$, where $\mathcal{I}$ is the unit tensor and $J \sim 130$ meV [2]. For isotropic coupling the spin-wave spectrum is doubly degenerate and the spin-wave energy $\omega(\mathbf{q})$ goes to zero as $q \to 0$. The existence of 3D long range antiferromagnetic (AF) order, with $T_{\rm N} \sim 250 - 400$ K, and the existence of some non-zero spin-wave gaps at $q = 0$ require an *anisotropic* exchange tensor in the plane, and/or some 3D exchange coupling. However, some of these small gaps, predicted by theory, have not yet been observed experimentally.

Although some of the phenomena in the cuprates can be explained by a classical treatment of anisotropic Heisenberg models which ignore quantum fluctuations, the cuprates still have several interactions which are *frustrated* at the mean-field level, and would lead to a ground state degeneracy. One example concerns the *in-plane exchange anisotropy* in the planar square lattice common to most tetragonal cuprates. Symmetry implies that the principal axes of $\mathbf{J}_{ij}$ in the plane are parallel to the $i - j$ bond ($\|$), perpendicular to the bond in the plane ($\perp$), and perpendicular to the plane (z). Thus, the spin interaction along this bond is of the form $J_\| S_{i\|} S_{j\|} + J_\perp S_{i\perp} S_{j\perp} + J_z S_{iz} S_{jz}$. Indeed, a five-band Hubbard model, in the limit when the on-site Coulomb repulsion dominates the hopping matrix elements [3], including both spin-orbit and Coulomb exchange interactions, yields deviations of the principal values J_α by a few parts in 10^4 from their average, J. The out-of-plane gap, $\omega_{\rm out} = 5$ meV, observed in many cuprates [2], is related to the out-of-plane anisotropy field, $H_{\rm A}^{\rm out} \equiv (2J_\perp + 2J_\| - 4J_z)S$:

$$h\nu \equiv \omega \equiv \sqrt{2H_{\rm E}H_{\rm A}} \equiv \sqrt{8JSH_{\rm A}}, \tag{1}$$

where h is Planck's constant. (In what follows, we give ν in units of GHz, and ω in meV: 1 meV$/h$ = 241.8 GHz.) Since $J_\perp \neq J_\|$, one might expect a similar gap, $\omega_{\rm in}$, for in-plane spin waves at $\mathbf{q} = 0$. However, for a classical spin model at $T = 0$, the sum over perpendicular bonds yields an isotropic planar energy and hence $\omega_{\rm in} = 0$. This isotropy is removed by *quantum fluctuations*, and detailed calculations yield an in-plane anisotropy field

$$H_{\rm A}^{\rm in,K} = 8K/S = C(J_\| - J_\perp)^2/J, \tag{2}$$

where $C \approx 0.16$ and the quantum four-fold energy per unit cell is $\mathcal{H}_4 = -K\cos 4\theta$ (θ is the angle between the staggered moment and a Cu–Cu bond, and there are two Cu ions per cell) [3]. $H_{\rm A}^{\rm in,K}/S$ is of order $1/S$ ($K \propto S$), emphasizing that $H_{\rm A}^{\rm in,K}$ is a manifestation of quantum fluctuations [4]. The fact that $H_{\rm A}^{\rm out}$ is linear in the exchange anisotropy, whereas $H_{\rm A}^{\rm in,K}$ is quadratic in that small quantity, indicates why the in-plane gap due to $H_{\rm A}^{\rm in,K}$ is too small to be detected by inelastic neutron scattering, thus requiring the present electron spin resonance (ESR) experiments.

A second (and more familiar) example of frustration occurs in materials like $Sr_2CuCl_2O_2$ ("2122"), which has the body centered tetragonal K_2NiF_4 structure with the CuO_2 layers in the c plane [6]; each Cu couples to four equidistant Cu's in a neighboring plane. For isotropic inter–plane exchange, the mean field sum of these four interactions vanishes. Nevertheless, below $T_{\rm N} \simeq 251$ K, the spins have a well defined AF structure, with the easy axis believed to be parallel to the [110] direction [7]. The magnetic structures of such cuprates have been explained [8] by considering, in addition to K, two competing energies, which also relieve the frustration. The first of these is an effective *bilinear interplanar coupling* [9] of the form $-j_{\rm eff}(\mathbf{S}_i \cdot \mathbf{S}_j)^2$ which is generated by *quantum fluctuations* of the otherwise frustrated isotropic interactions. This effective coupling favors colinearity of the spins in neighboring planes. The second additional energy, $A_{\rm pdip}$, arises from the small interplanar "pseudodipolar" exchange anisotropic interaction (not yet calculated), which adds to the dipolar interaction $A_{\rm dip}$, giving a contribution,

$$H_{\rm A}^{\rm in,d} \equiv 4A/S \equiv 4(A_{\rm dip} + A_{\rm pdip})/S, \tag{3}$$

to the in-plane anisotropy, where the parameter A was defined in Ref. [8].

$Sr_2Cu_3O_4Cl_2$ ("2342") combines the above quantum effects. In 2342, the CuO planes have an additional Cu ion (denoted CuII) at the center of every alternate CuI plaquette. The CuI subsystem shows AF ordering at $T_{\rm NI} \simeq 380$ K. Although the isotropic molecular field acting on the CuII sites from the CuI's vanishes (similar to the interplanar field in 2122), the CuII subsystem exhibits a small ferromagnetic moment below $T_{\rm NI}$, and shows AF ordering at $T_{\rm NII} \simeq 40$ K, with its staggered moment colinear to that of the CuI's, i. e. along [110] (parallel to a CuI–CuI bond). Both the magnetic structure [10,11] and spin-wave spectrum [5] have been explained by including all the three mechanisms mentioned above.

Here we report on ESR measurements of the in–plane fluctuation induced gap in both 2342 and 2122 [12]. The observation of these small gaps in the predicted range of energy gives decisive confirmation both of the model, in which the gaps are attributed (at least partly) to quantum fluctuations, and also of our fundamental understanding of the electronic structure of the cuprates which is used to calculate J_α. Also, the magnetic field dependence of the gap gives values for the anisotropic g tensor for the Cu spins, which roughly agree with theory.

The single crystals of 2122 and 2342 used in this study were grown at MIT by slow cooling from the melt. The ESR measurements were performed using the spectrometer installed in RIKEN [13]. Several Klystrons and Gunn oscillators were used to cover the frequency range from 20 to 100 GHz. We also used a vector network analyzer, bought from the AB millimetre Company, operating in the frequency range 50-700 GHz. Magnetic fields up to 20 T were generated with a superconducting magnet from Oxford Instruments. Since the ESR signals in these samples were weak at low frequencies, we used resonant cavities at respective frequencies below 70 GHz and a field modulation technique to enhance the sensitivity.

Figure 1 shows typical ESR signals, observed at 70 K in a single crystal of 2342 with the external magnetic field (**H**) parallel to the easy axis [110] at the designated frequencies. Because we use different resonant cavities for different frequencies, a direct comparison of the absorption intensity is difficult. However, we see a tendency of the intensity (which is obtained by a double integration of the spectrum with respect to H) to decrease with decreasing frequency. The intensity in the 2122 sample is much weaker than that of the 2342 sample.

Since 2342 has two transitions, we measure the antiferromagnetic resonance (AFMR) at two temperatures: 70 K, between $T_{\rm NI}$ and $T_{\rm NII}$, and 1.5 K ($< T_{\rm NII}$). In Figs. 2 and 3 we plot the frequency (ν) dependence of the resonance fields observed in single crystals of 2342 and 2122, at the indicated temperatures and field directions. The experimental points constitute separate branches, each showing clearly an energy gap at H=0. Each set of data has been fitted by [14]

$$\nu(H)^2 = \nu(0)^2 + (g\mu_B H/h)^2, \tag{4}$$

where g is the g value for the corresponding orientation of $\mathbf{H}$, and $\mu_{\rm B}$ the Bohr magneton. The fitted coefficients, with their statistical errors, are given in Table I.

Now we compare the experimental results with the theory, beginning with 2342 at 70 K. Ignoring the small ferromagnetic moment (which only introduces a negligible shift in the effective K), the intermediate phase ($T_{\rm NII} < T < T_{\rm NI}$) is similar to the "usual" AF phase of other cuprates. The AF ordering of the CuI's generates only two low energy modes, $\omega_{\rm out}$ and $\omega_{\rm in}$, given by Eq. (1) with $H_{\rm A}^{\rm out}$ and $H_{\rm A}^{\rm in,K}$. The former has an energy of order 5 meV, which is too large for our AFMR measurements (but has been measured by neutrons in Ref. [5]). Using the experimental value of $\omega_{\rm in}$ at 70 K in Eqs. (1) and (2) gives $K = \omega_{\rm in}^2/(64J) = (5.2 \pm 0.3) \times 10^{-7}$ meV [4]. This roughly agrees with the static experimental value of Kastner *et al.* [11], $K = (10 \pm 3) \times 10^{-7}$ meV. Returning to Eq. (2), these two values of K imply that $\delta J \equiv |J_\| - J_\perp| = \sqrt{8JK/(SC)} \approx 0.08 - 0.11$ meV, where we use J=130 meV [5] and $S = 1/2$ [4]. This value of δJ is a factor of two larger than that estimated theoretically by Entin-Wohlman *et al.* [15], for the geometry of 2122. A larger value of δJ for 2342, compared to 2122, could result from the difference in environment (due to insertion of CuII's), from the uncertainties in the Hubbard model parameters used in Refs. [3,15], or from higher order renormalizations [4]. Unlike the other gaps discussed below, $\omega_{\rm in}$ at 70 K for 2342 is purely due to fluctuations, which are of quantum origin at $T = 0$; $H_{\rm A}^{\rm in,d} = 0$ for the CuI spins (which sit on top of each other in neighboring planes). Thus, this measurement presents a clear confirmation of the theory involving $\mathcal{H}_4$.

Before continuing with 2342, we now turn to 2122. Unlike 2342 above $T_{\rm NII}$, where minimizing $\mathcal{H}_4$ causes the spins to point along the CuI–CuI bond, in 2122 the spins are believed to point colinearly along [110], i. e. at 45^o with the Cu-Cu bond [7]. This implies that $H_{\rm A}^{\rm in,K}$ from Eq. (2) enters into the in-plane gap with a *negative* sign, and that the actual gap must also have positive contributions from the interplanar exchange anisotropies, which dominate $\mathcal{H}_4$. Our analysis [16] indeed yields

$$\begin{aligned}\omega_{\rm in}^2 &= 2H_{\rm E}(H_{\rm A}^{\rm in,d} - H_{\rm A}^{\rm in,K}) \\ &= 32J(A_{\rm dip} + A_{\rm pdip}) - 8JSH_{\rm A}^{\rm in,K} \ , \end{aligned} \qquad (5)$$

where we used Eq. (3). We now show that this equation is reasonably fulfilled. We take $\delta J = 0.04$ meV from Ref. [15], and thereby get $H_{\rm A}^{\rm in,K} = 2 \times 10^{-6}$ meV. We also take $A_{\rm dip} \approx 2.7 \times 10^{-6}$ meV [17]. The experimental value $\omega_{\rm in} \approx 0.048$ meV then implies that $A_{\rm pdip} \approx -2\times10^{-6}$ meV. Writing $A_{\rm pdip} = 2S^2\delta J_{\rm int}$, where $\delta J_{\rm int}$ is the anisotropy of the interlayer exchange interaction [8], we find $\delta J_{\rm int} \sim -10^{-5}$ meV. Assuming that $|\delta J_{\rm int}| \sim 10^{-4} J_{\rm int}$, we estimate an interplanar exchange energy $J_{\rm int} \sim 0.1$ meV, of the same order of magnitude as the estimate for La_2CuO_4, $J_{\rm int} \approx 0.25$ meV [8]. Thus we have corroborated Eq. (5).

We now return to the low$-T$ phase ($T<T_{\rm NII}$) in 2342. This phase has six spins per unit cell, and four low energy modes [5], of which two, denoted in Ref. [5] by ω_3 and ω_4, with energies above 5 meV, were used there to measure the parameter $j_{\rm eff}$ related to the I–II biquadratic coupling. The two new modes at lower frequency, which we call $\omega_{\rm in}^<$ and $\omega_{\rm out}^<$, represent respectively in-plane and out-of-plane fluctuations, mostly on the CuII ions (these were denoted ω_1 and ω_2 in Ref. [5]). $\omega_{\rm out}^<$ concerns out-of-plane fluctuations of the CuII spins. These are practically not affected by $H_{\rm A}^{\rm in,K}$, and $\omega_{\rm out}^<$ is equal to ω_2 in Kim *et al.* [5]. Using the parameters as listed in [5], we predict $\omega_{\rm out}^< = 1.77$ meV, which agrees nicely with the present result 422.5 GHz (=1.747 meV).

The effective in-plane anisotropy energy was neglected in the theoretical expressions in Ref. [5], because it had only insignificant effects on the modes studied there. In contrast, $\omega_{\rm in}^<$ is determined by this energy. Accordingly we use Eq. (1), but now we have $H_{\rm E} = 4SJ_{\rm II}$, where $J_{\rm II} \approx 10.5$ meV is the CuII-CuII exchange energy [10], and $H_{\rm A}^{\rm in}$ has contributions from *both* CuI and CuII. Our new spin wave analysis yields [16]

$$H_{\rm A}^{\rm in} = (8k_{\rm eff}/S)J/(J - J_{\rm I-II} + 2J_{\rm II}) \ , \qquad (6)$$

where $J_{\rm I-II} = -10$ meV [18] and the relevant anisotropies are contained in the parameter $k_{\rm eff}$, where

$$k_{\rm eff} = k + \tfrac{1}{2}A - K_{\rm II}. \qquad (7)$$

The first term, k, contains the four-fold anisotropy energy of the CuI's, and contributions from the ferromagnetic canting of both the CuI and CuII. Unlike the higher T phase, the canting of the CuII is now not negligible, and we recover [11]

$$k = 2K + 8J_{pd}^2 M_{\rm I}^{\dagger 2}[0.53/(8J_{\rm II})] \qquad (8)$$

($M_{\rm I}^\dagger$ is the staggered moment on the CuI's, and J_{pd} is the pseudodipolar part of the CuI–CuII exchange). The last two terms in Eq. (7) come from the CuII spins. Since the spin structure of the CuII ions is similar to that of the Cu's in 2122 (the spins point at 45^o to the CuII-CuII bond), these two terms are analogous to those in Eq. (5). Using Eq. (2), with $|J_\parallel - J_\perp| \sim 10^{-4}J$ for the CuII's, we estimate that $K_{\rm II} \sim K/10$. At low T, $k_{\rm eff}$ is thus dominated by the interplanar dipolar term $\frac{1}{2}A$. Assuming for simplicity only real dipolar interactions, we have $A = 3(g\mu_B M_{\rm II}^\dagger)^2 X$, where X is the lattice sum in Eq. (10) of Ref. [8], which we evaluated as $7\times10^{-4}\mathring{A}^{-3}$. Thus, at $T = 0$ we estimate $k_{\rm eff} \approx 24\times10^{-6}$ meV, in reasonable agreement with the static value [11]. The mysterious dramatic increase in $k_{\rm eff}$ observed in Ref. [11] below $T_{\rm NII}$ is thus explained by the additional dominant term $\frac{1}{2}A$ (which is proportional to $M_{\rm II}^{\dagger 2}$). Using this estimate in Eq. (6) yields $\omega_{\rm in}^< \approx 0.12$ meV, not far from the experimental value 0.15 meV.

The g-tensor is calculated from $\mathcal{H} = \mu_B \mathbf{H}\cdot(\mathbf{L}+2\mathbf{S})$. The quantum average is calculated for the ground state, so one has $g = 2+g_L$, where $<\mathbf{L}>= g_L <\mathbf{S}>$. The latter is calculated perturbatively, with the spin-orbit term $\lambda\mathbf{L}\cdot\mathbf{S}$, i. e.

$$\begin{aligned}< L_\alpha > &= < L_\alpha(1/E)\lambda\mathbf{L}\cdot\mathbf{S} > + < \lambda\mathbf{L}\cdot\mathbf{S}(1/E)L_\alpha > \\ &= 2\lambda < S_\alpha >< L_\alpha(1/E)L_\alpha >, \end{aligned} \qquad (9)$$

where E represents the energy of an intermediate state.

Thus,

$$\begin{aligned} g_{x,y} &= 2 + 2\lambda/e_{x,y} \\ g_z &= 2 + 8\lambda/e_z. \end{aligned} \qquad (10)$$

Taking $e_x = e_y = e_z = 1.8$ eV and $\lambda = 0.1$ eV gives $g_{x,y} = 2.11$, $g_z = 2.45$, in reasonable agreement with the present results 2.08 and 2.30.

In conclusion, we have measured the low energy excitations of the $S{=}\frac{1}{2}$ tetragonal antiferromagnets, $Sr_2CuO_2Cl_2$ and $Sr_2Cu_3O_4Cl_2$ using an ESR spectrometer covering a wide range of frequency and magnetic field. At 70 K for 2342, we have been successful in observing the quantum in-plane energy gap at $H{=}0$ predicted theoretically. The other in-plane gaps which we have measured reflect additional anisotropies, which we have shown to have the expected orders of magnitude.

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[1] J. G. Bednorz and K. A. Müller, Z. Phys. B**64**, 189 (1986).

[2] M. A. Kastner, R. J. Birgeneau, G. Shirane and Y. Endoh, Rev. Mod. Phys. **70**, 897 (1998).

[3] T. Yildirim, A. B. Harris, A. Aharony and O. Entin-Wohlman, Phys. Rev. B**52**, 10239 (1995).

[4] As discused in Ref. [5], theoretical expansions in powers of $1/S$ usually imply some quantum renormalization factors for ω. Since Eq. (2) already represents a quantum correction, we have not included these factors. However, this might imply an uncertainty in the theoretical values, which could involve a factor of up to 2.

[5] Y. J. Kim, A. Aharony, R. J. Birgeneau, F. C. Chou, O. Entin-Wohlman, R. W. Erwin, M. Greven, A. B. Harris, M. A. Kastner, I. Ya. Korenblit, Y. S. Lee and G. Shirane, Phys. Rev. Lett. **83**, 852 (1999).

[6] Von B. Grande and Hk. M*ü*ller-Buschbaum, Z. Anorg. Allg. Chem. **417**, 68 (1975).

[7] D. Vaknin, S. K. Sinha, C. Stassis, L. L. Miller and D. C. Johnston, Phys. Rev B **41**, 1926 (1990).

[8] T. Yildirim, A. B. Harris, O. Entin-Wohlman and A. Aharony, Phys. Rev. Lett. **72**, 3710 (1994).

[9] E. F. Shender, Sov. Phys. JETP **56**, 178 (1982).

[10] F. C. Chou, A. Aharony, R. J. Birgeneau, O. Entin-Wohlman, M. Greven, A. B. Harris, M. A. Kastner, Y. J. Kim, D. S. Kleinberg, Y. S. Lee and Q. Zhu, Phys. Rev. Lett. **78**, 535 (1997).

[11] M. A. Kastner, A. Aharony, R. J. Birgeneau, F. C. Chou, O. Entin-Wohlman, M. Greven, A. B. Harris, Y. J. Kim, Y. S. Lee, M. E. Parks and Q. Zhu, Phys. Rev. B **59**, 14702 (1999).

[12] The ESR experiment [H. Ohta, M. Sumikawa, M. Motokawa, S. Noro and T. Yamadaya, J. Phys. Soc. Jpn. **64**, 1759 (1995)] made on a similar compound $Ba_2Cu_3O_4Cl_2$ did not reveal a clear in-plane gap, because data points were not sufficient.

[13] M. Hagiwara, K. Katsumata, I. Yamada and H. Suzuki, J. Phys.: Condens. Matter **8**, 7349 (1996).

[14] T. Nagamiya, K. Yosida and R. Kubo, Adv. Phys. **4**, 1 (1955).

[15] O. Entin-Wohlman, A. B. Harris, and A. Aharony, Phys. Rev. B**53**, 11661 (1996).

[16] Details will be published elsewhere.

[17] In Ref. [8] the constant $A_{\rm dip}$ was evaluated using $S = 0.2$ as mentioned in their Ref. 18. However, we prefer to use $S = 0.3$. Since $A_{\rm dip}$ is proportional to S^2, we set $A_{\rm dip} = 1.2 \times 10^{-6}(0.3/0.2)^2$ meV.

[18] $J_{\rm I-II}$ is known to be ferromagnetic from S. Tornow, O. Entin-Wohlman and A. Aharony, Phys. Rev. B**60**, 10206 (1999). Its magnitude was estimated in Ref. [5].

TABLE I. Experimental results.

	T, K	**H** ∥	g	$\nu(0)$, GHz	ω, meV
2342	70	[110]	2.083(3)	16.0(9)	$\omega_{\rm in} = 0.066(4)$
2342	1.5	[110]	2.080(2)	36.1(6)	$\omega_{\rm in}^{<} = 0.149(3)$
2342	1.5	[001]	2.301(1)	422.5(1)	$\omega_{\rm out}^{<} = 1.7473(4)$
2122	5	[110]	2.046(2)	11.6(7)	$\omega_{\rm in} = 0.048(3)$

FIG. 1. The electron spin resonance signal observed in a single crystal of $Sr_2Cu_3O_4Cl_2$ at 70 K and at several frequencies. **H** ∥ [110].

FIG. 2. The frequency versus magnetic field plots of the ESR signals observed in a single crystal of $Sr_2Cu_3O_4Cl_2$ at 1.5 K for the two field directions, and at 70 K for **H** ∥ [110]. Inset: same data, ν^2 versus H^2.

FIG. 3. The frequency dependence of the resonance field in a single crystal of $Sr_2CuO_2Cl_2$ obtained at 5 K for **H** ∥ [110]. The dotted line is drawn through the origin, for comparison. Inset: same data, ν^2 versus H^2.

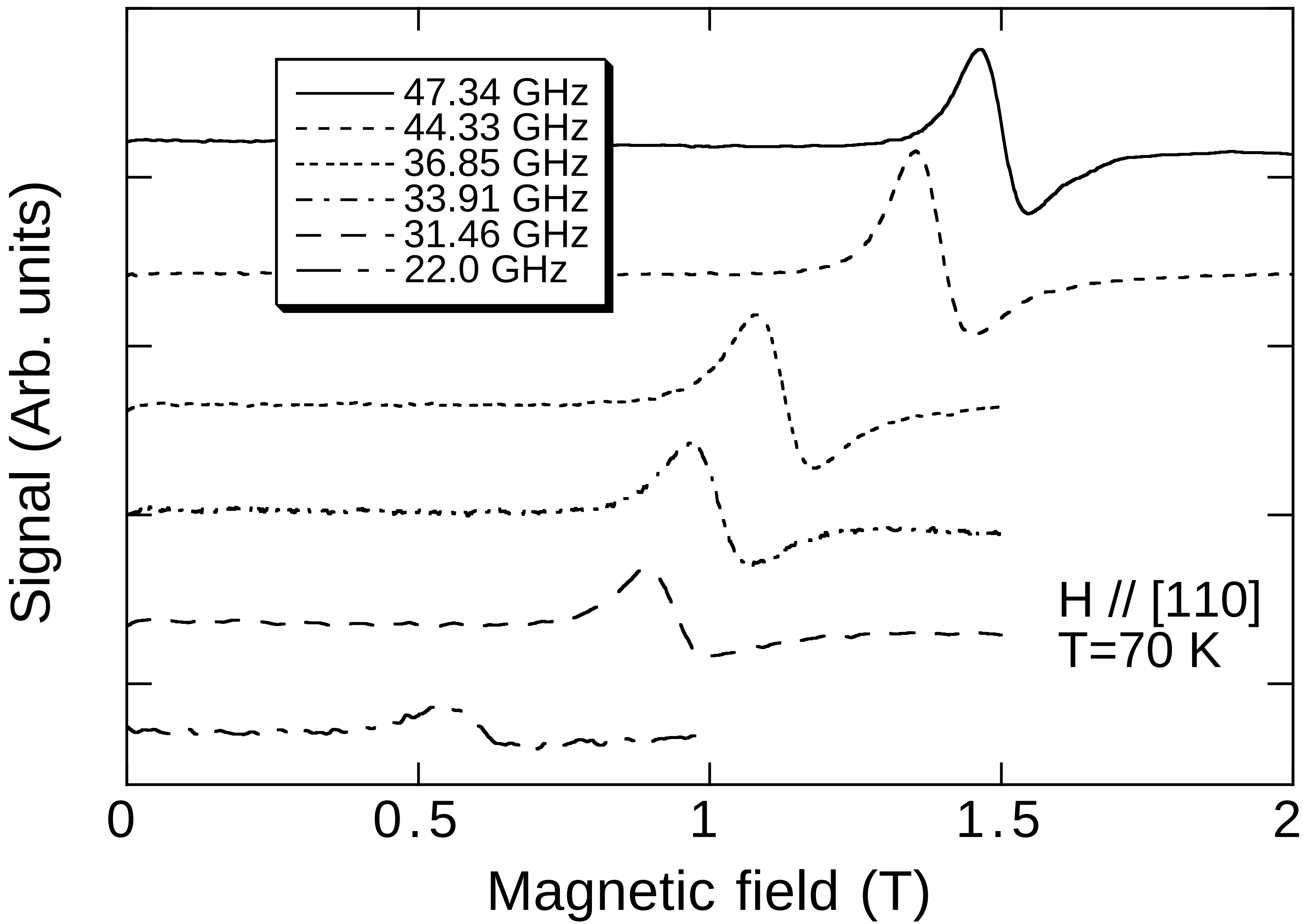

Fig.1 Katsumata et al.

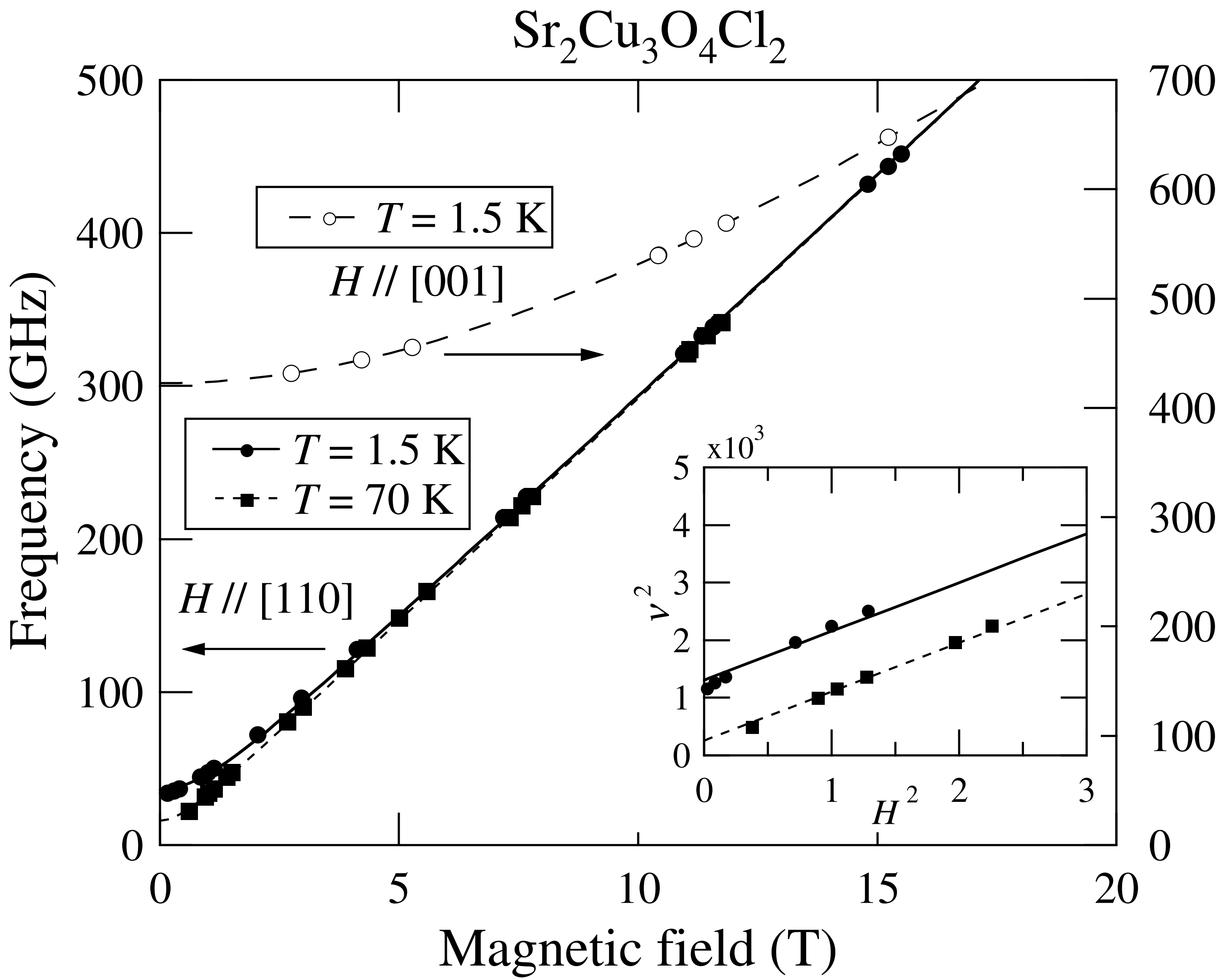

Fig.2 Katsumata *et al.*

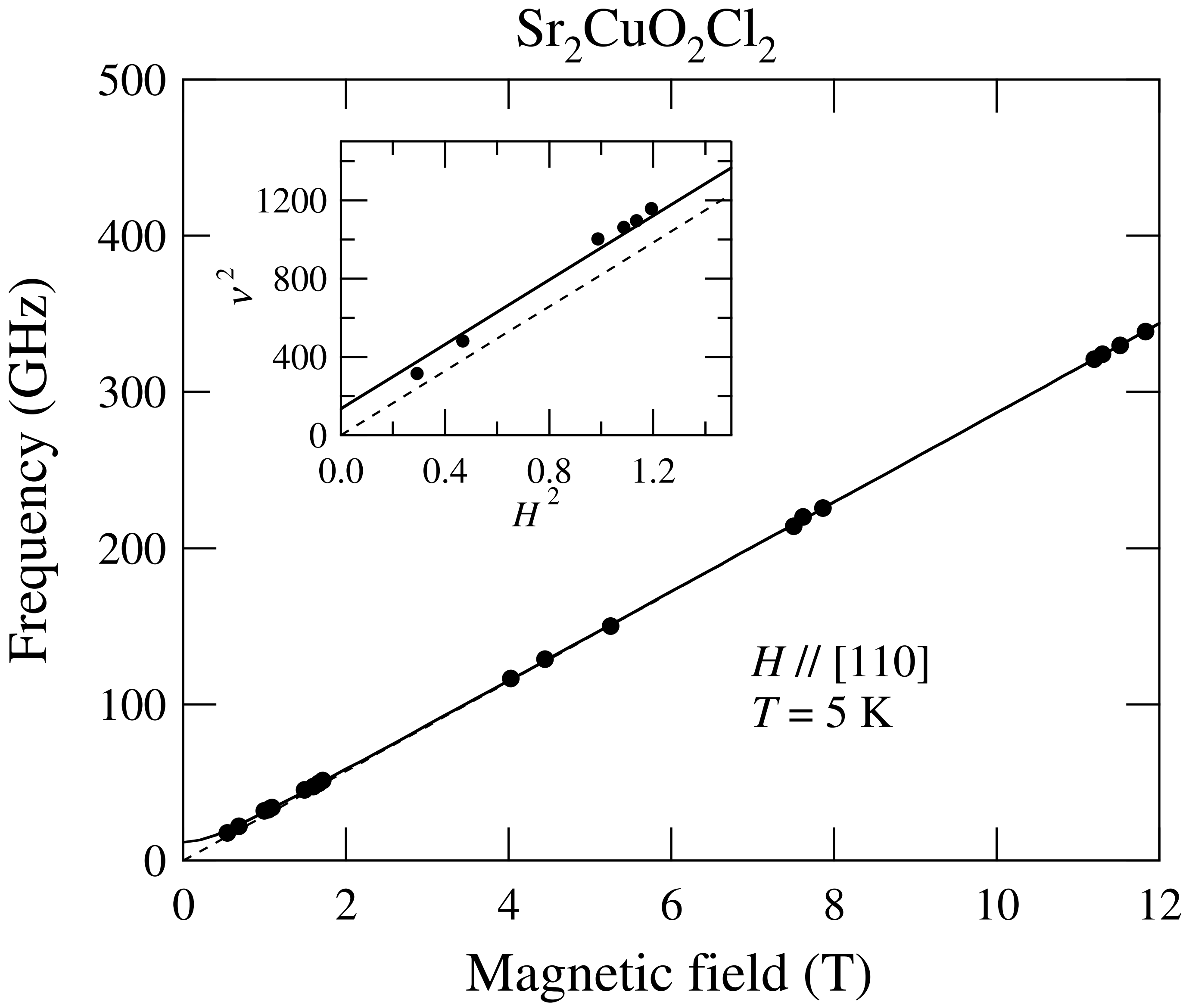

Fig.3 Katsumata *et al.*