High Field ESR on LiCu$_2$O$_2$

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Instrument: Magnet & Spectrometer

Sample:

Incident light always along \( a \)

External field can point parallel or perpendicular to plane: along \( a \), or \( b \) or \( c \)

Polarization: along \( a \) or \( c \)

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Brookhaven Lab, NSLS, U12IR

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Spectrometer Magnet, Detector

14/16 Tesla

8 - 200 cm\(^{-1}\)
(Up to visible)

1.8K-300K

Transmission
ESR: magnetic field dependent optical absorption, $q = 0$ magnon gap

- $\text{CuSO}_4$: “paramagnetic” standard (Mihaly)
- $\text{LaMnO}_3$: antiferromagnet $T_N$=135K (Jianshi Zhu, U. Texas)
- $\text{NaNiO}_2$: triangular spin array (DeBrion, GHMFL)
- $\text{LiCu}_2\text{O}_2$: spin $\frac{1}{2}$ chain $T_c$=24K (Berger/Forró, EPFL)
- $\text{Ni}_5(\text{TeO}_3)_4\text{Cl}_2$ spin 1 planar $T_c$=23K (Berger/Forró, EPFL)
- $\text{BaCu}_2\text{Si}_2\text{O}_7$, $\text{BaCu}_2\text{Ge}_2\text{O}_7$, $\text{BaCu}_2\text{SiGeO}_7$: $T_c$~9K (Zheludev, ORNL)

- $\text{Mn}_{12}$-acetate: effective spin 10, large anisotropy (Tu, Sarachik, CUNY, Y23.00007)
- Dimitri Basov (UC San Diego), Andrei Sirenko (NJIT), Adrain Gozar (BNL) ...
LiCu$_2$O$_2$

two copper sites, spin $\frac{1}{2}$ Cu$^{++}$ ions make triangular ladder

A.M. Vorotynov et al. JETP 86, 1020 (1998)
A.A. Gippius et al., PRB 70, 020406 (2004)
T. Masuda et al., PRL, 92, 177201 (2004);PRB, 72, 014405 (2005)
LiCu₂O₂

From Masuda et al., PRL 92, 177201 (2004)

Order: q=(0.5, 0.174, 0.5)
Frustrated exchange coupling

\[ \mathcal{H} = \sum_{i,j} J_1 S_{i,j} S_{i+1,j} + J_2 S_{i,j} S_{i+2,j} + J_4 S_{i,j} S_{i+4,j} + J_{\perp} S_{i,j} S_{i,j+1} - g\mu_B H S_{i,j}^y + \mathcal{H}' \]

- \( J_1 = 6.4 \text{meV} \)
- \( J_2 = -11.9 \text{meV} \) (ferromagnetic.)
- \( J_4 = 7.4 \text{meV} \)
- \( J_{\perp} = 1.8 \text{meV} \)

From neutron scattering


Frustration --> helical spin order.

\( J \)-s are not enough! What selects the plane of the helix? How large is the gap at \( q=0 \) in the magnon spectrum?

Exchange anisotropy

\[ \mathcal{H}' = D_{\perp} S_{i,j}^y S_{i+1,j}^y \]

ESR provides the answer
Temperature dependence

Field dependence at several temperatures

LiCu$_2$O$_2$ transmission relative to 0T

Transmission ratio

Wavenumber (cm$^{-1}$)

Temperature dependence at fixed field (12T)

Transmission ratio

Wavenumber (cm$^{-1}$)

Temperature dependence at fixed fields

Paramagnetic state: \( g=2.3 \)  Signal disappears around transition temperature, recovered at higher frequency at low T.
Field dependence at low temperature

Field applied perpendicular to plane of helix.
Zero field: Spin gap of 11.5cm⁻¹=1.4meV
Average of three data set  
- three main findings

- Zero field gap:
  \[ \Delta = 11.5 \text{cm}^{-1} = 1.4 \text{meV} \]

- Field dependence:
  \[ \omega \propto \sqrt{H^2 + H_0^2} \]

- Absorption depends strongly on field
Contrast to: LaMnO$_3$

$\text{Ni}_5(\text{TeO}_3)_4\text{Cl}_2$

L. Mihály, T. Fehér, B. Náfrádi, H. Berger
L. Forró, to be published

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Field dependence: theory

Helical order with canting


\[ J' = \frac{J(2Q) + J(0) - J(Q)}{4} \]

Zero field gap:

\[ \Delta = 2S \sqrt{J' \cos(\phi/2) D_{ex}} \]

Two branches.
ESR susceptibility is strongly field dependent, weak signal for upper branch

LiCu$_2$O$_2$ Conclusions

Spin gap at $q = 0$: $\Delta = 11.5 \text{cm}^{-1} = 1.4 \text{meV}$

Exchange anisotropy: $D_{\text{ex}} \cos (\phi/2) = 0.079 \text{meV}$
with $\phi = 5.2 \text{ rad}$.

Estimates of quantum fluctuations (Anderson): $\Delta S = 0.125$
$S = 1/2$ $\rightarrow$ $S_{\text{eff}} = 0.375$

Reduced $g$ factor of $g = 1.8$