Title: Dynamics of the magnetic susceptibility deep in the Coulomb phase of the dipolar spin ice material Ho2Ti2O7

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Abstract: Very recently, it has been recognized that excitations out of the ground state of materials known as spin ice can be viewed as magnetic monopoles, the magnetic analog to electric charges. Like electrons and positrons,
these particles possess a charge of +Q or -Q and therefore attract or repel each other. Magnetic monopoles, however, can be accelerated using a magnetic field instead of an electric field. In this talk, I will report on
experiments deep into the frozen state of the spin ice material holmium titanate where monopoles are few and far between and the material responds very slowly to a changing magnetic field. Taking advantage of the
extremely sensitive magnetic field detector known as a superconducting quantum interference device (SQUID), we measure the rate at which the monopoles are created, move about and are eventually annihilated. A surprisingly simple law emerges at low temperatures, known as an Arrhenius law, suggesting that the generation of these magnetic charges requires an energy that does not change in temperature and for a yet unknown reason, is precisely 3 times the energy required to make a single, bare monopole.

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AC Susceptibility Measurements of $Ho_2Ti_2O_7$

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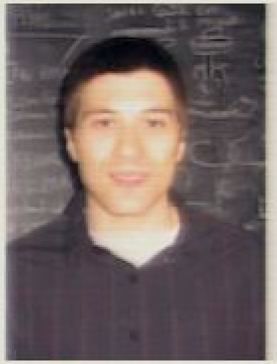


Borko
Djurkovic

Halle Revell

Motivation

- Recent work by Jaubert and Holdsworth interpret measurements by Snyder et al. to have evidence of monopole-like excitations in spin ices.
- Two-in two-out maps to a vacuum
- Three-in one-out (three-out one-in) maps to a quasi-particle (monopole)
- Dipolar interaction between spins maps to Coulomb interactions between monopoles
- Magnetic charges behave as a Coulomb gas of monopoles.

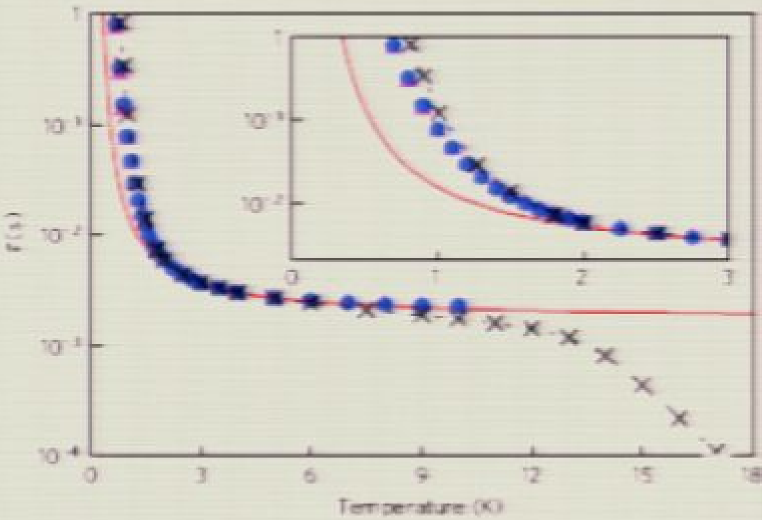


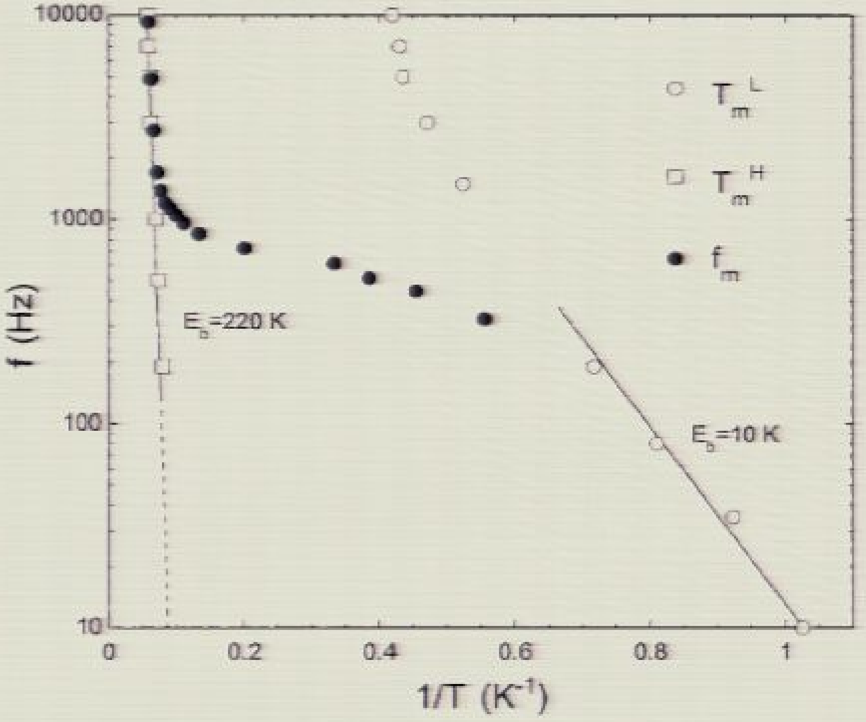
Figure 2 | Relaxation timescales τ in $Dy_2Ti_2O_7$: experiment and simulation. The experimental data (crosses) are from Snyder et al.[3] Th
Arrhenius law (red line) represents the free diffusion of topological defe
in the nearest-neighbour model. The relaxation timescale of the Dirac
string network driven by Metropolis dynamics of magnetic monopoles h
been obtained for fixed chemical potential (pink filled triangles) and wit
varying slowly to match the defect concentration in dipolar spin ice (blu
filled circles). The temperature scale is fixed without any free parameter
Inset: The same data shown in the low-temperature region.

Spin Ice:
Ramirez, A. Hayashi, R. Cava, R. Siddharthan, and B. Shastry, Nature (London) **399**, 333 (1999).
C. den Hertog and M. J. P. Gingras, Phys. Rev. Lett. **84**, 3430 (2000).
J. Gardner, M. Gingras, and J. E. Greedan, Rev. Mod. Phys. **(2010)**

Matsuhira, Hinatsu, Sakakibara
Journal of Physics, Condensed Matter (2001)

Powder sample

Snyder *et al* have somewhat similar parallel result
Nature 2001



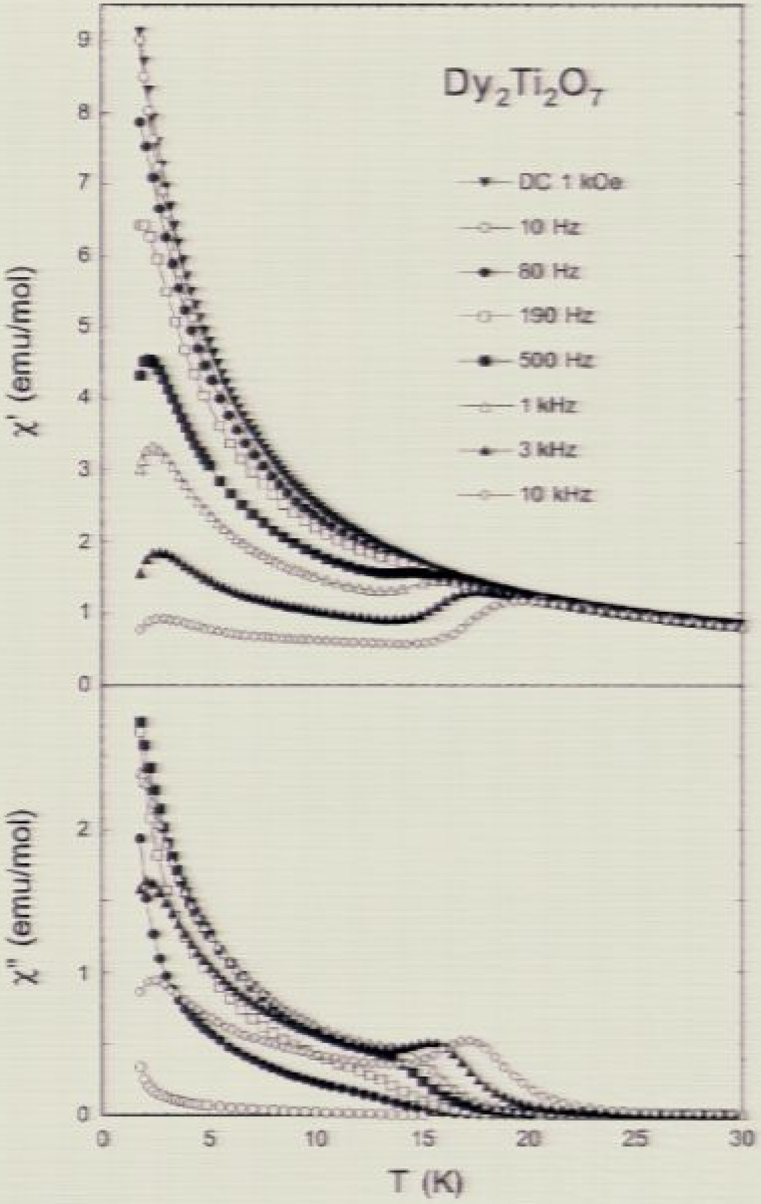
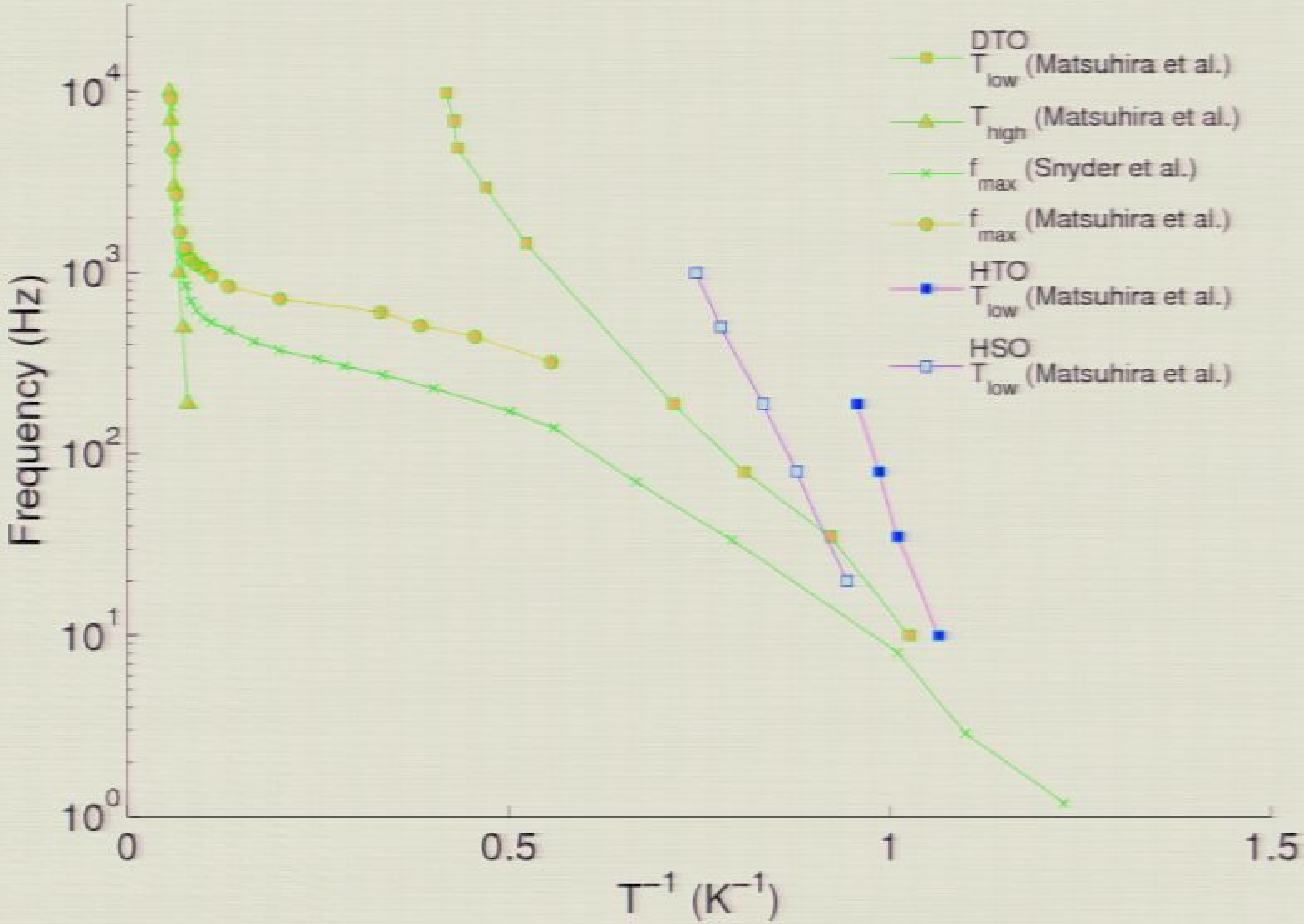


Figure 2. Temperature dependence of χ' and χ'' of $Dy_2Ti_2O_7$ above 1.8 K.


Frequency (Hz)
10^4
10^3
10^2
10^1
10^0
0
0.5
1
1.5
T^{-1} (K^{-1})
DTO
T_{low} (Matsuhira et al.)
T_{high} (Matsuhira et al.)
f_{max} (Snyder et al.)
f_{max} (Matsuhira et al.)
HTO
T_{low} (Matsuhira et al.)
HSO
T_{low} (Matsuhira et al.)

Conventional Susceptometer

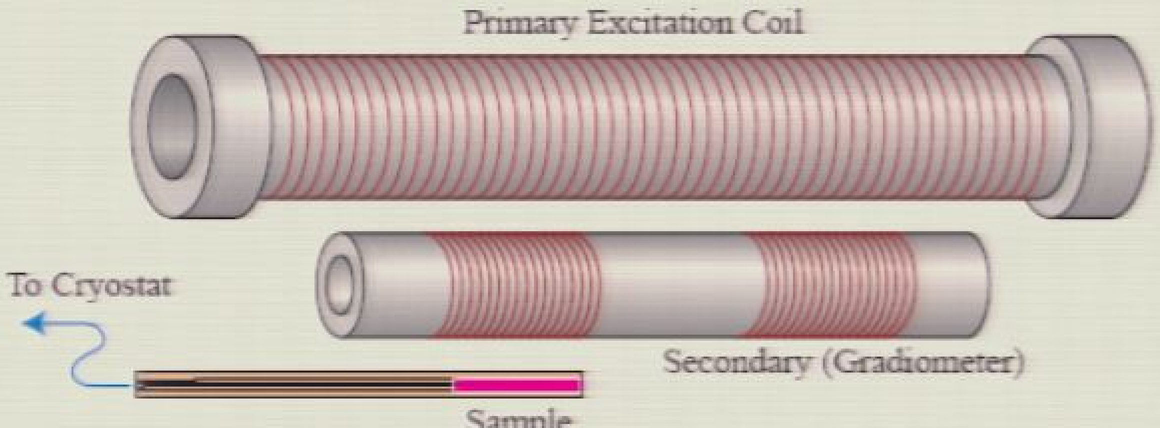
Advantage: Easy to put together and use.

Disadvantages:
Loses sensitivity at low frequencies since signal is due to induced EMF.

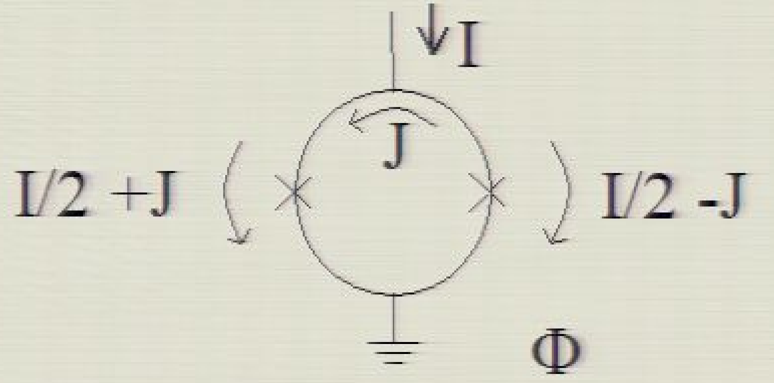
$$V_{EMF} \propto \frac{d\phi}{dt} \propto \omega$$

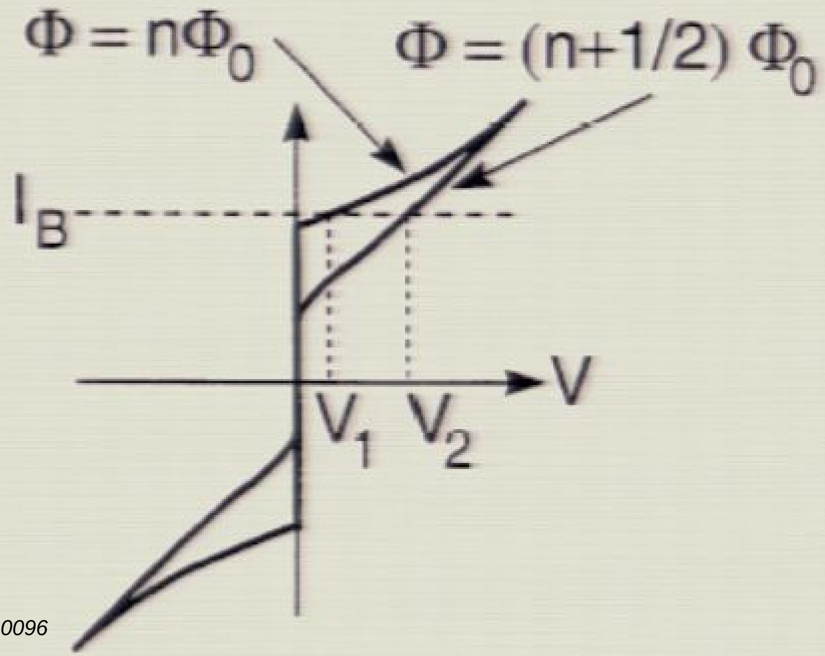
Too many turns reduces highest useable frequency due to intercoil resonance.

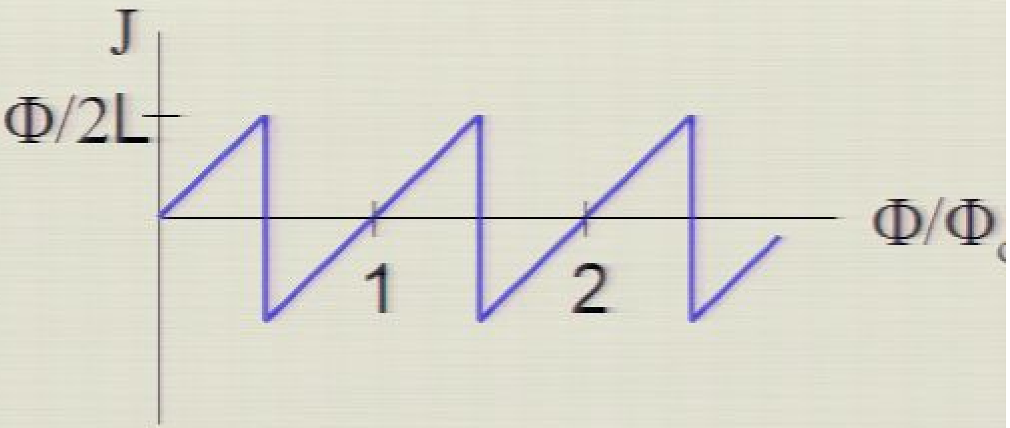
---Phase shifts and non-flat frequency response.

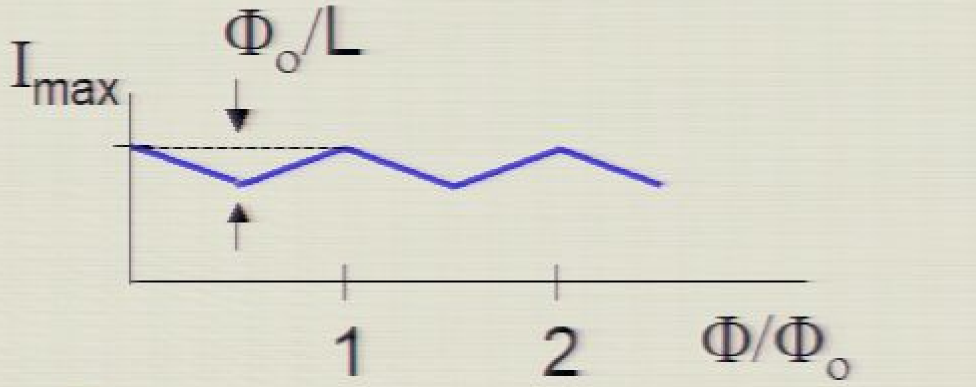


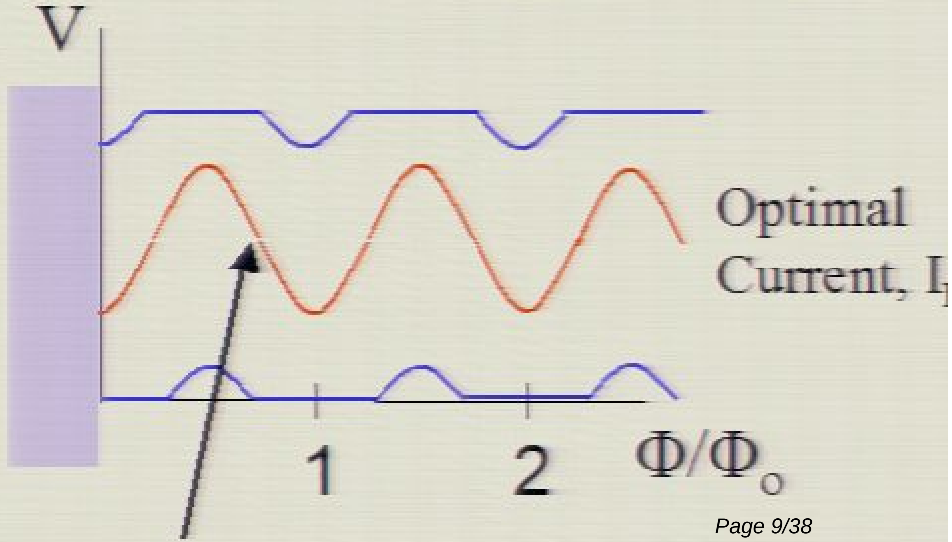
The DC SQUID
Sensitive flux to voltage converter











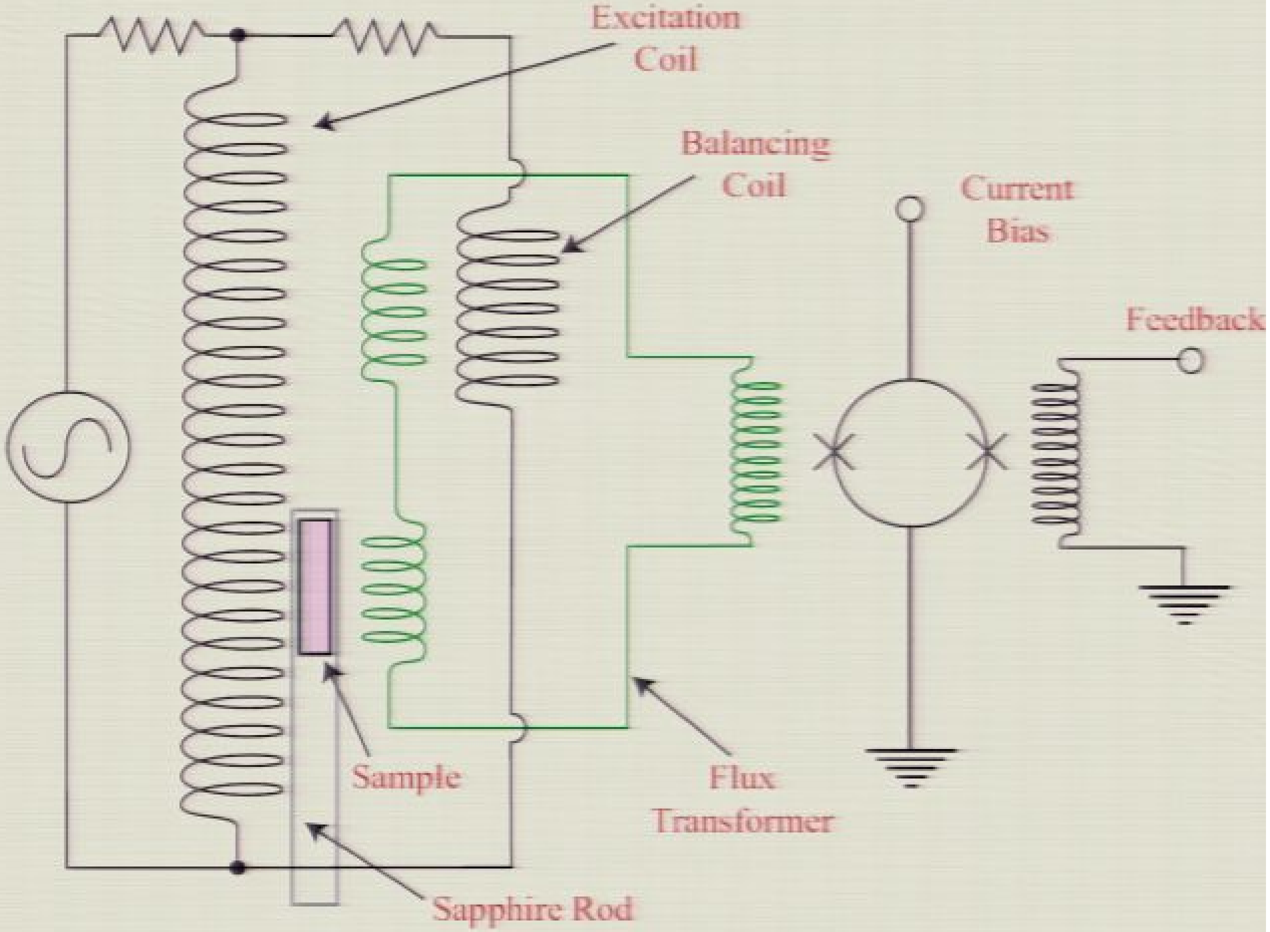
Sensitivity $\sim 1\mu\phi/\sqrt{Hz}$

Optimal operating point

SQUID Magnetometer Measurement

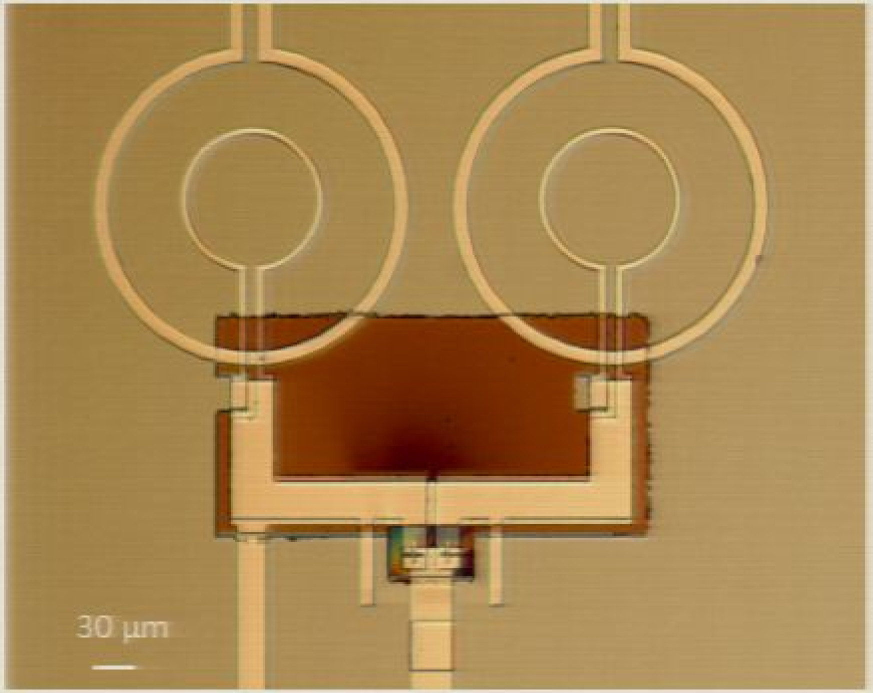
- Use a SQUID with a superconducting flux transformer to make a magnetometer.
- The current sent to the feedback coil produces an equal and opposite field to that provided by the flux transformer.
- This device directly measures flux, as opposed to induced EMF. **Flat Frequency response. No problems with phase shifts**.

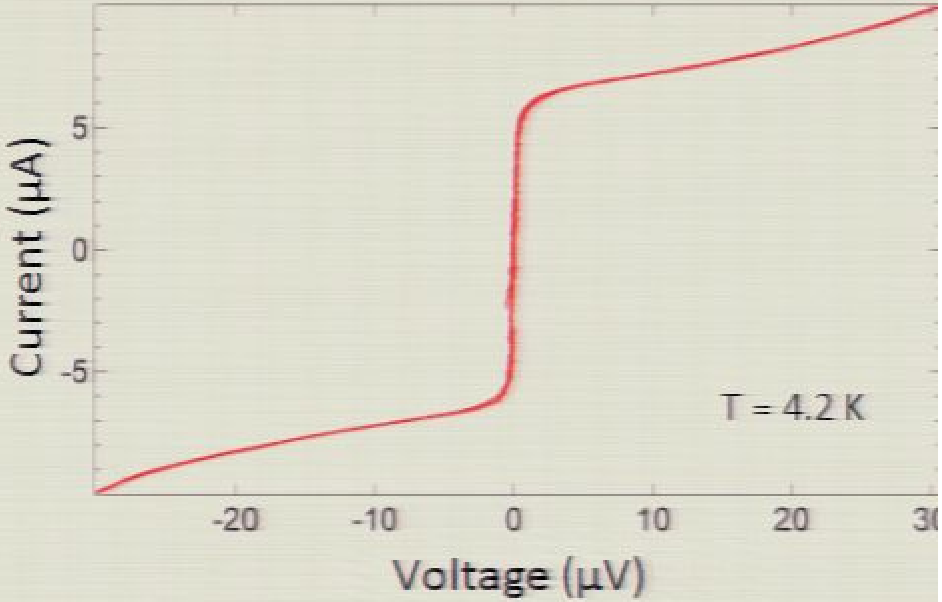
SQUID Magnetometer

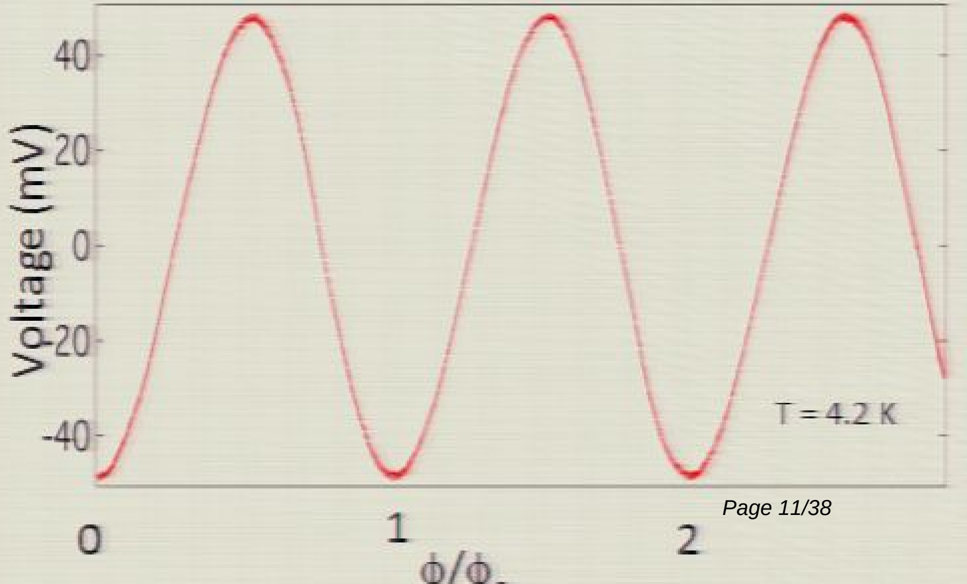


SQUID and controller from ez-SQUID

Fabricating our own customized Nb SQUIDS







Advantages:

- Should work in relatively high field
- Simple demagnetization correction
- Can be multiplexed for multiple measurements at the same time.

Spin Ice

- A spin ice – orders to degenerate ground state analogous to that of water ice
- Spin ices found so far: $Dy_2Ti_2O_7$ (DTO), $Ho_2Ti_2O_7$ (HTO), $Ho_2Sn_2O_7$, $Pr_2Sn_2O_7$
- Axial moments – ice rules, two-in two-out
- Well described by Dipolar Spin Ice Model
- $J_{eff} \cong 1.1K$ for DTO
- $J_{eff} \cong 1.8K$ for HTO

$$\mathcal{H} = -J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + D\, r_{nn}^3 \sum_{i>j} \left[\frac{\mathbf{S}_i \cdot \mathbf{S}_j}{|\mathbf{r}_{ij}|^3} - \frac{3\,(\mathbf{S}_i \cdot \mathbf{r}_{ij})\,(\mathbf{S}_j \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5} \right]$$

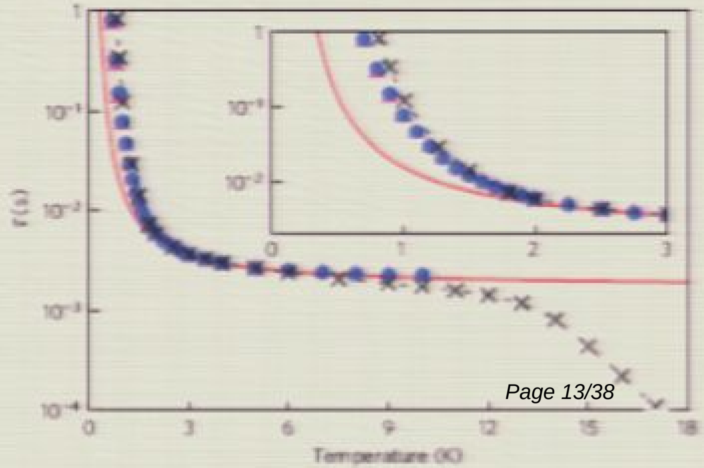
$$\mathcal{H} = -3J_{\mathrm{eff}} \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j = J_{\mathrm{eff}} \sum_{\langle i,j \rangle} \sigma_i\, \sigma_j \qquad J_{eff} = m^2 \frac{(5D - J)}{3}$$

	D	D_{nn}	J	J_{nn}	J_{eff}
Dy [20]	1.41	2.35	−3.72	−1.24	≈ 1.1
HoTi [21]	1.41	2.35	−1.65	−0.52	≈ 1.8

Temperature Regimes

- Three general relaxation regimes found in all measurements
- High T (~>15K)
  - Arrhenius with activation energy ~300K
  - On the order of separation to the next excited crystal field level
- Intermediate T (2K ~< T ~< 15K)
  - Almost temperature independent quantum tunnelling regime
  - High defect population (four in or four out, three in (out) three out (in))
  - At low end, monopole population becomes smaller.
  - Important energy scale: $2J_{eff}$
- Low T (~<2K)
  - Spins freezing out, drastic increase in relaxation time
  - Two in two out, difficult to form monopoles

$Dy_2Ti_2O_7$



Jaubert and Holdsworth

Snyder et al.

Demagnetization Correction

- Not used by all groups
- Our method:
  - Measure two sample geometries.
  - To find N, using analytical form for rectangular prism, given i Aharoni (J. Appl. Phys., 1998).

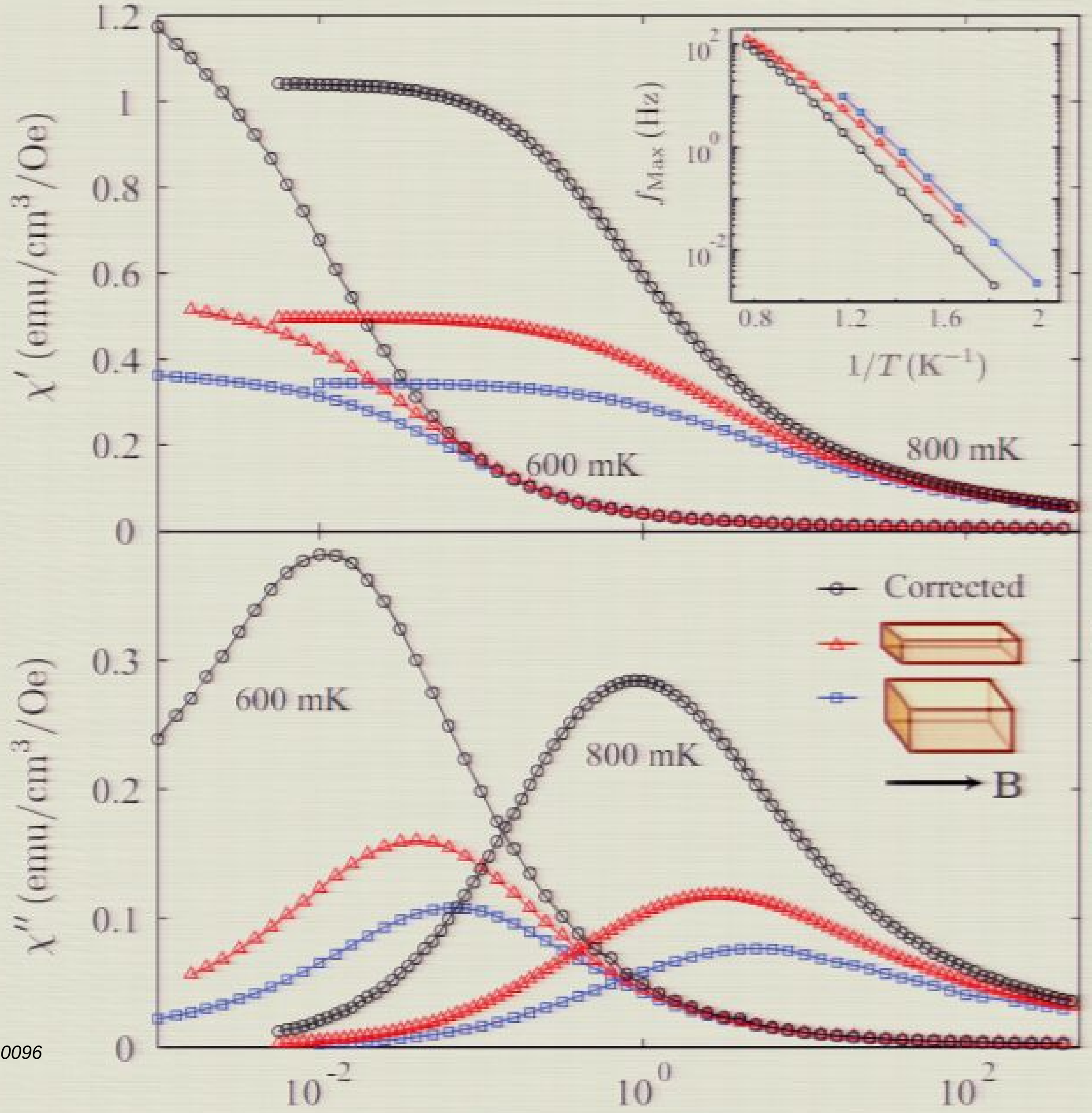
$$\chi = \chi' - i\chi'' \qquad \frac{1}{\chi} = \frac{1}{\chi_A} - 4\pi N$$

$$\chi' = \frac{\chi'_A - 4\pi N\left(\chi'^2_A + \chi''^2_A\right)}{(1 - 4\pi N\chi'_A)^2 + (4\pi N\chi''_A)^2},$$

$$\chi'' = \frac{\chi''_A}{(1 - 4\pi N\chi'_A)^2 + (4\pi N\chi''_A)^2}$$

  - Scale data sets by a calibration factor to find agreement between sets.
- Shifts peak absorption frequency lower, especially at low T.
- Demagnetization makes things seem slower than they actually are in true bulk ac susceptibility

Demagnetization Correction



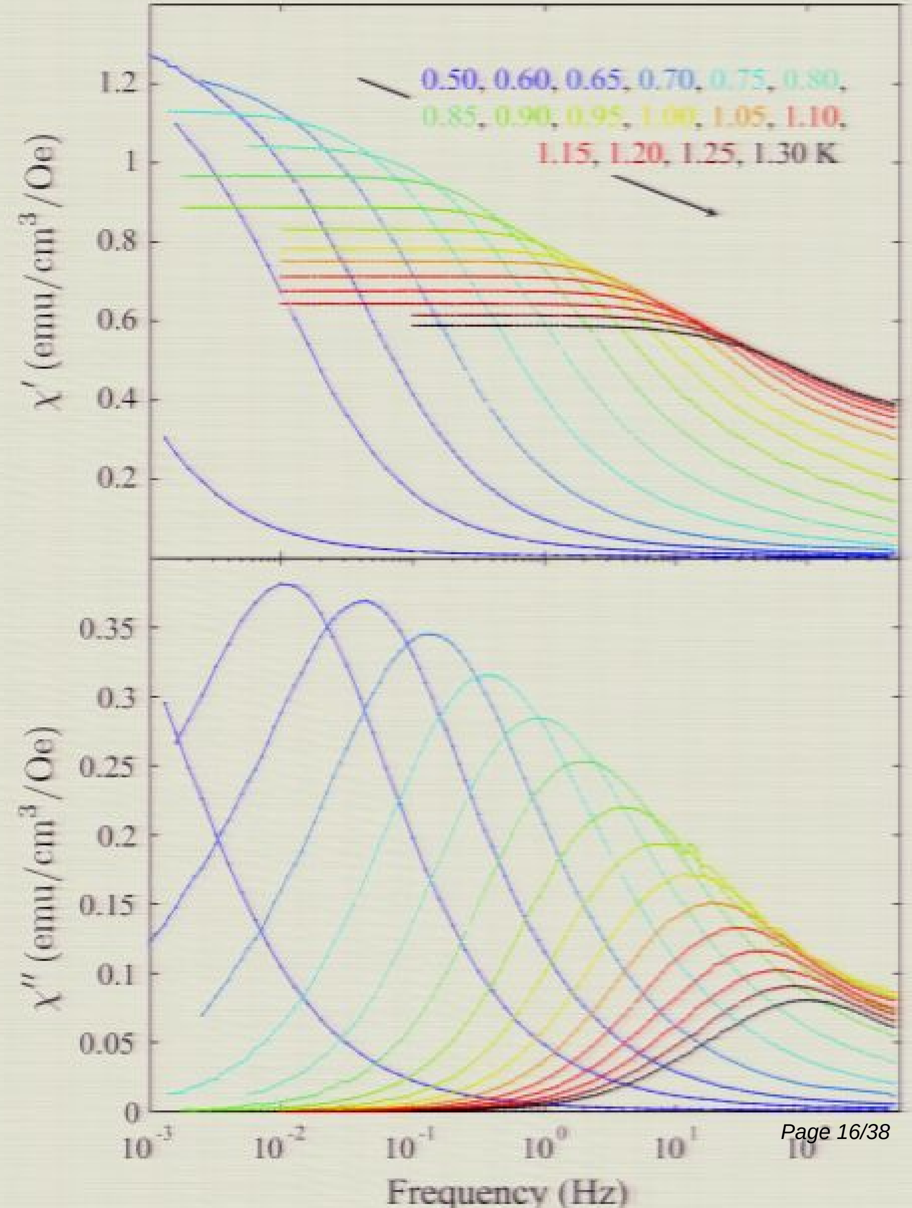
0.6mm x 0.6mm x 3mm

1.1mm x 1.1mm x 2.6mm

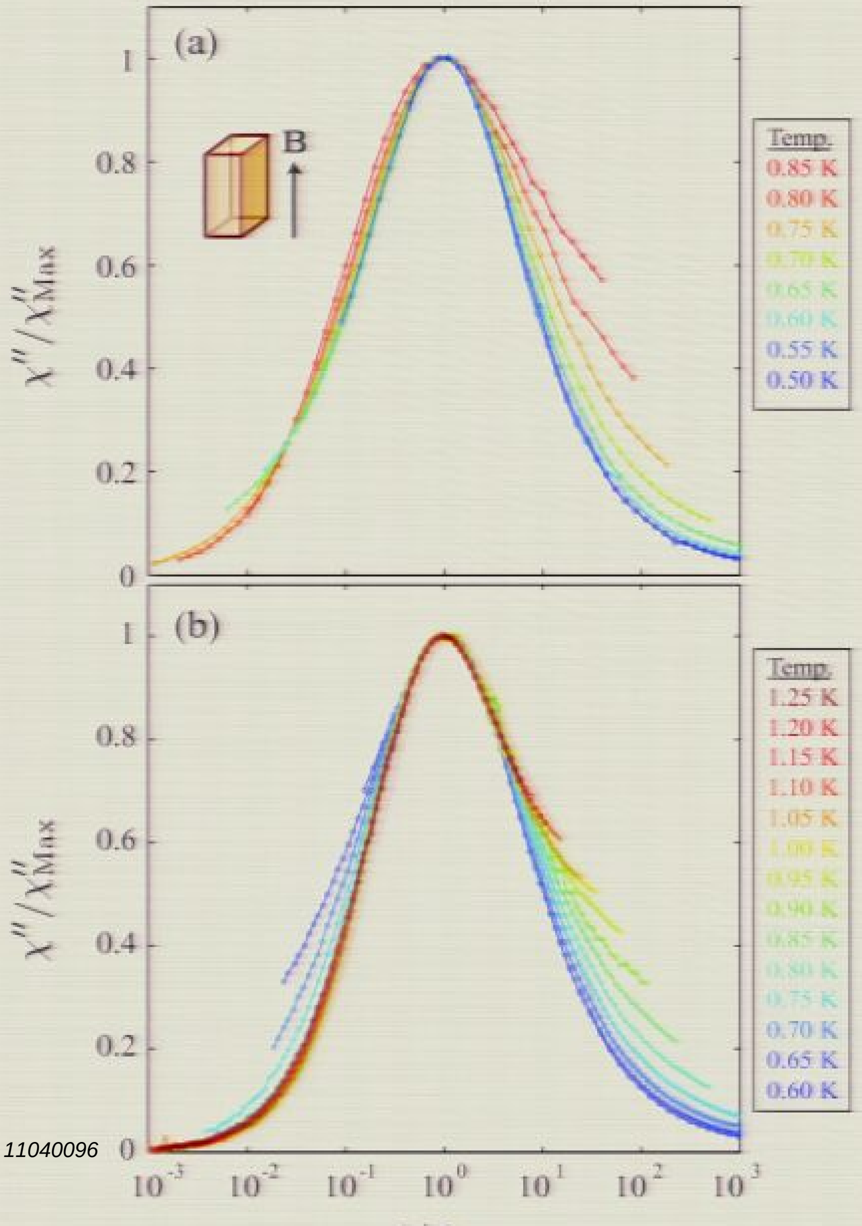
[110] along length

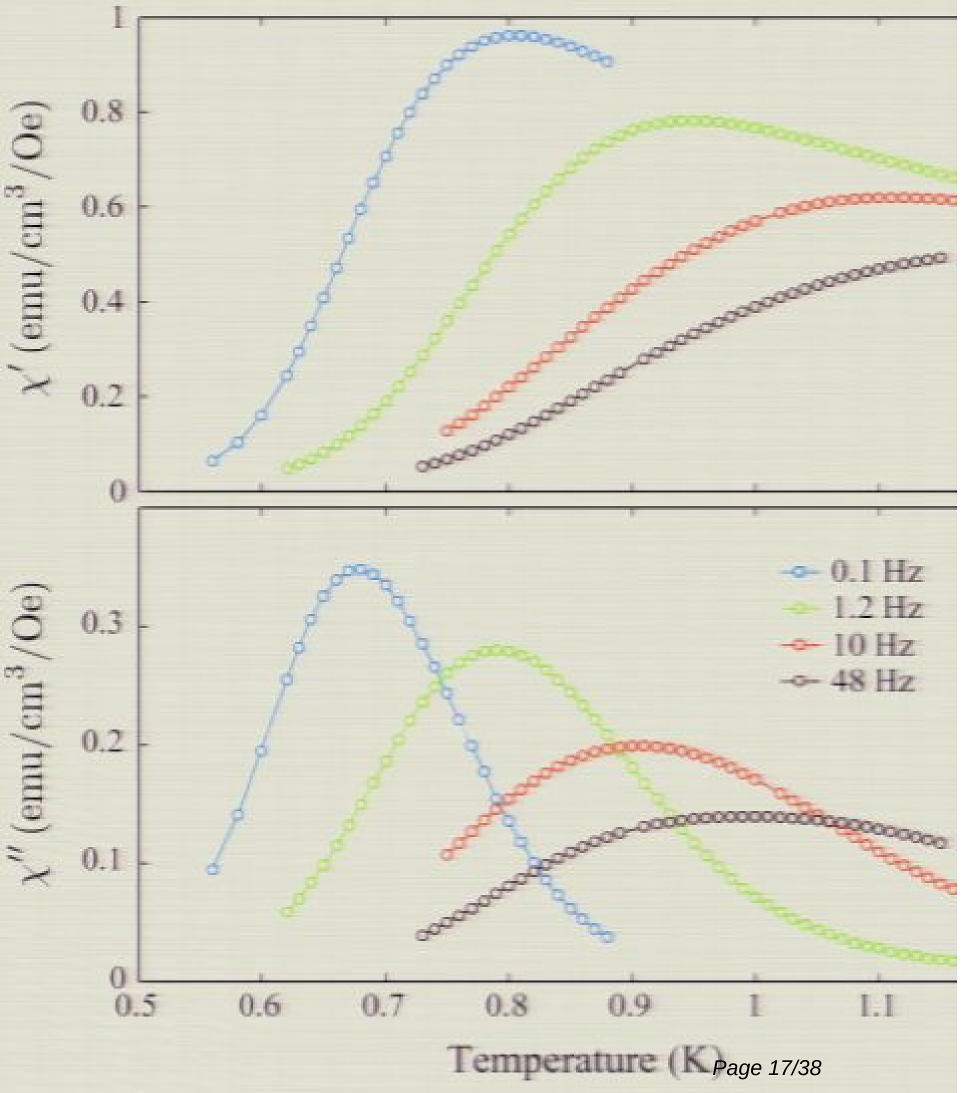
$Ho_2Ti_2O_7$ Results

- Corrected for demagnetization
- N = 0.084, 0.171
- Temperature increasing from blue to red



Absorption Spectra and Temperature Scans



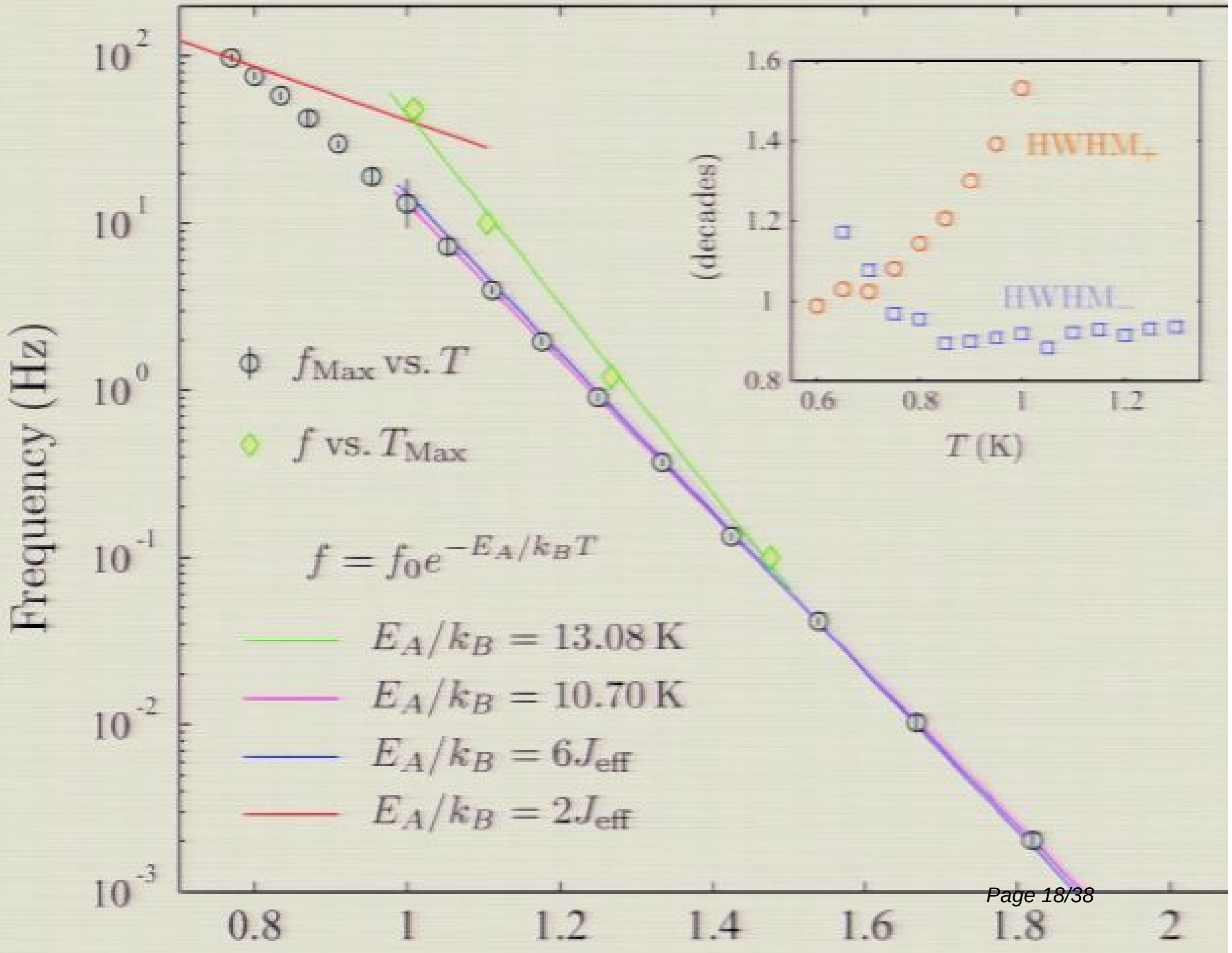


$Ho_2Ti_2O_7$ Results

- Arhennius law fits well for lower temperatures

$$\tau = \tau_0 \exp\left(\frac{E_A}{T}\right)$$

$\tau_0 = 1.74 \times 10^{-6}$ sec
$E_A/k_B = 10.7\ \mathrm{K} \pm 0.15\mathrm{K}$



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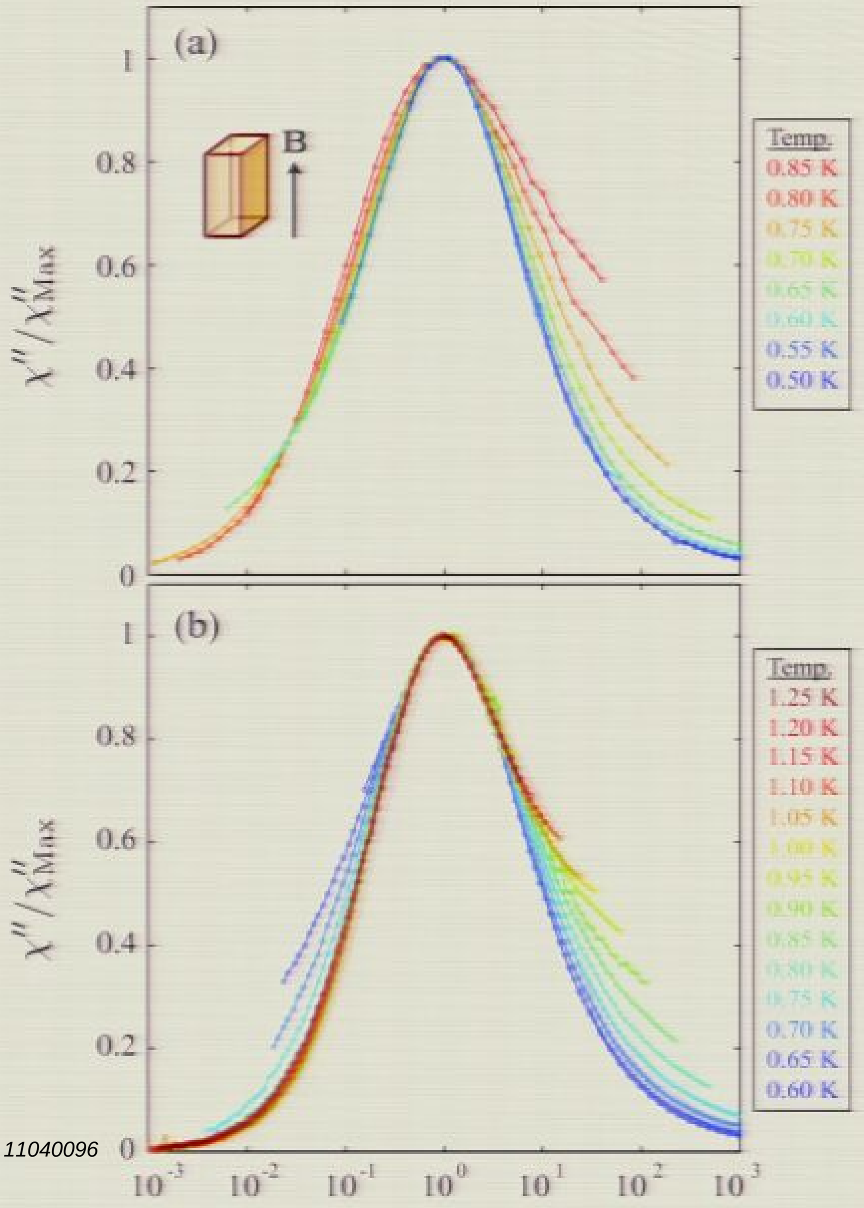

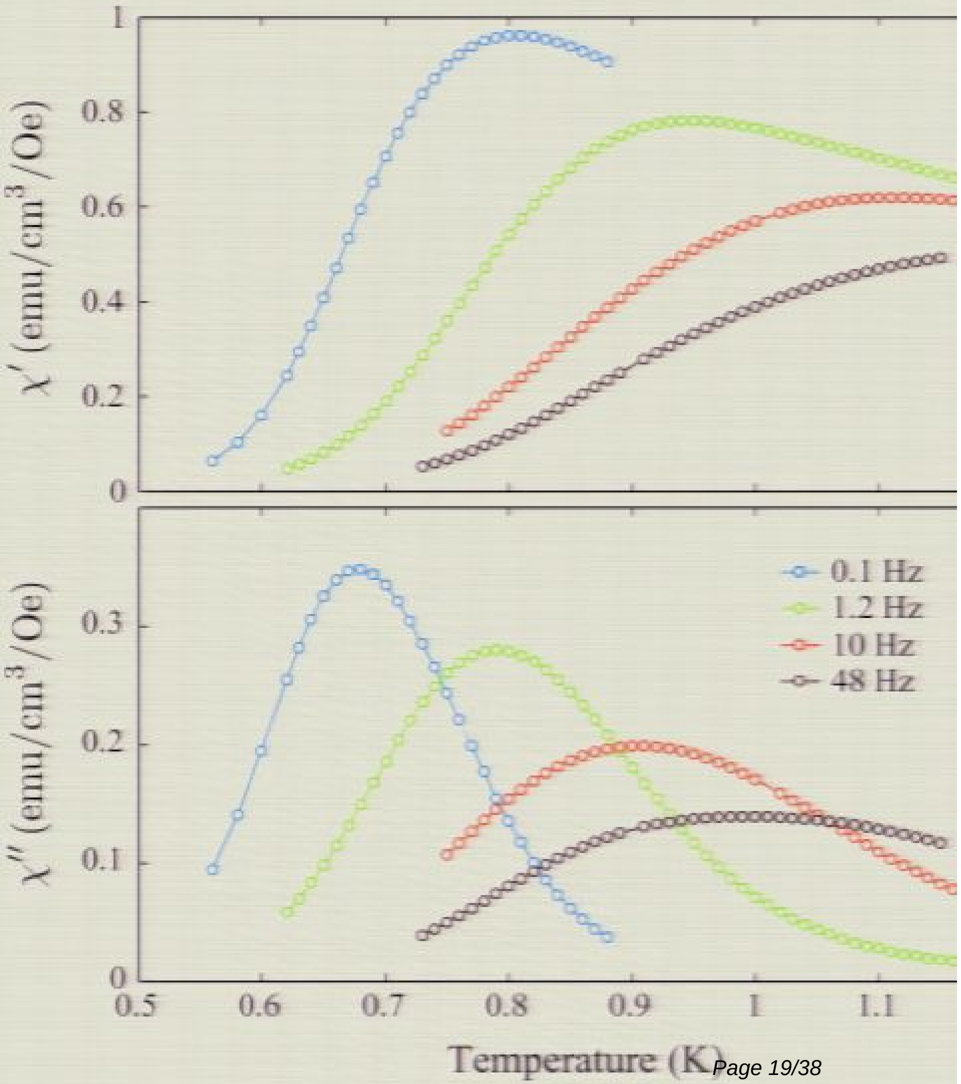

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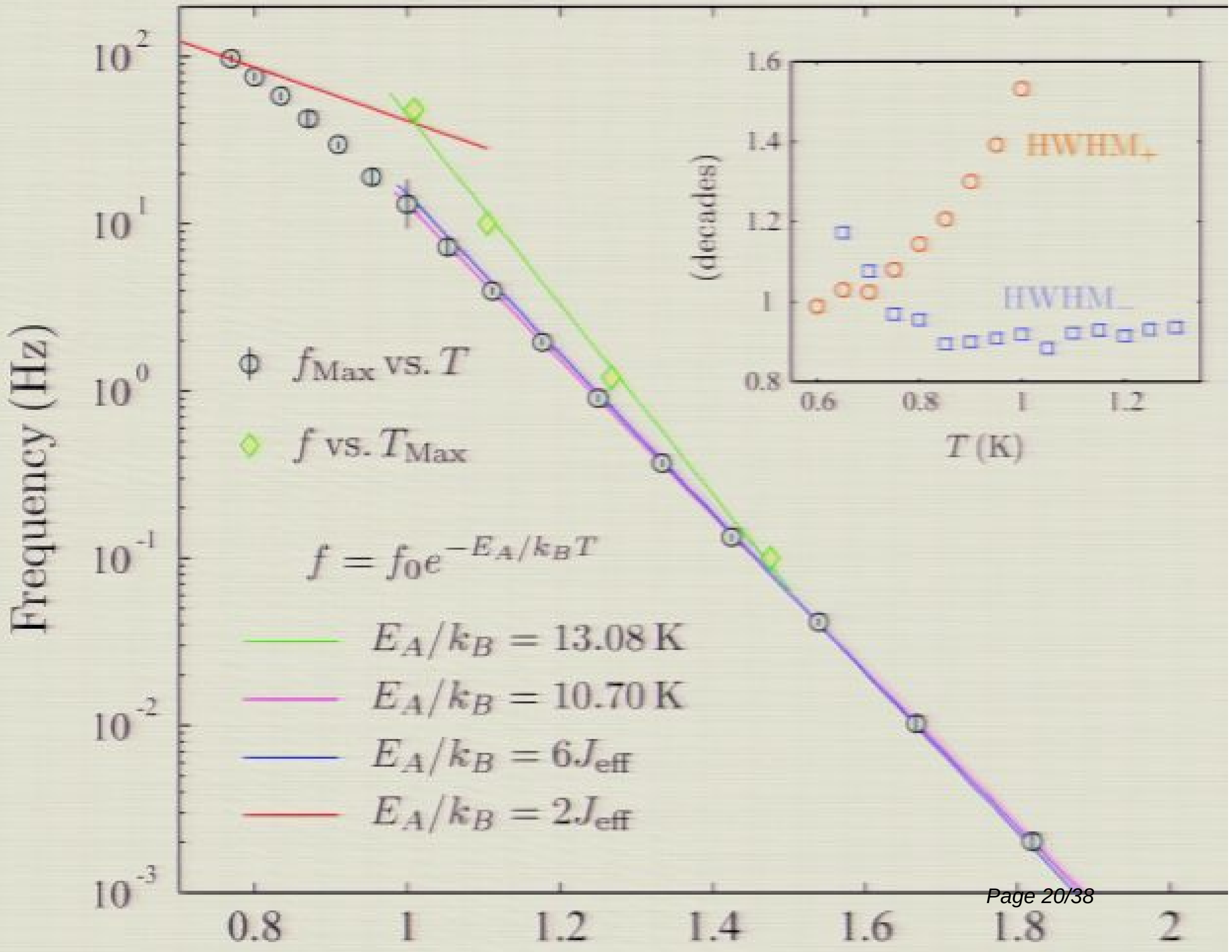
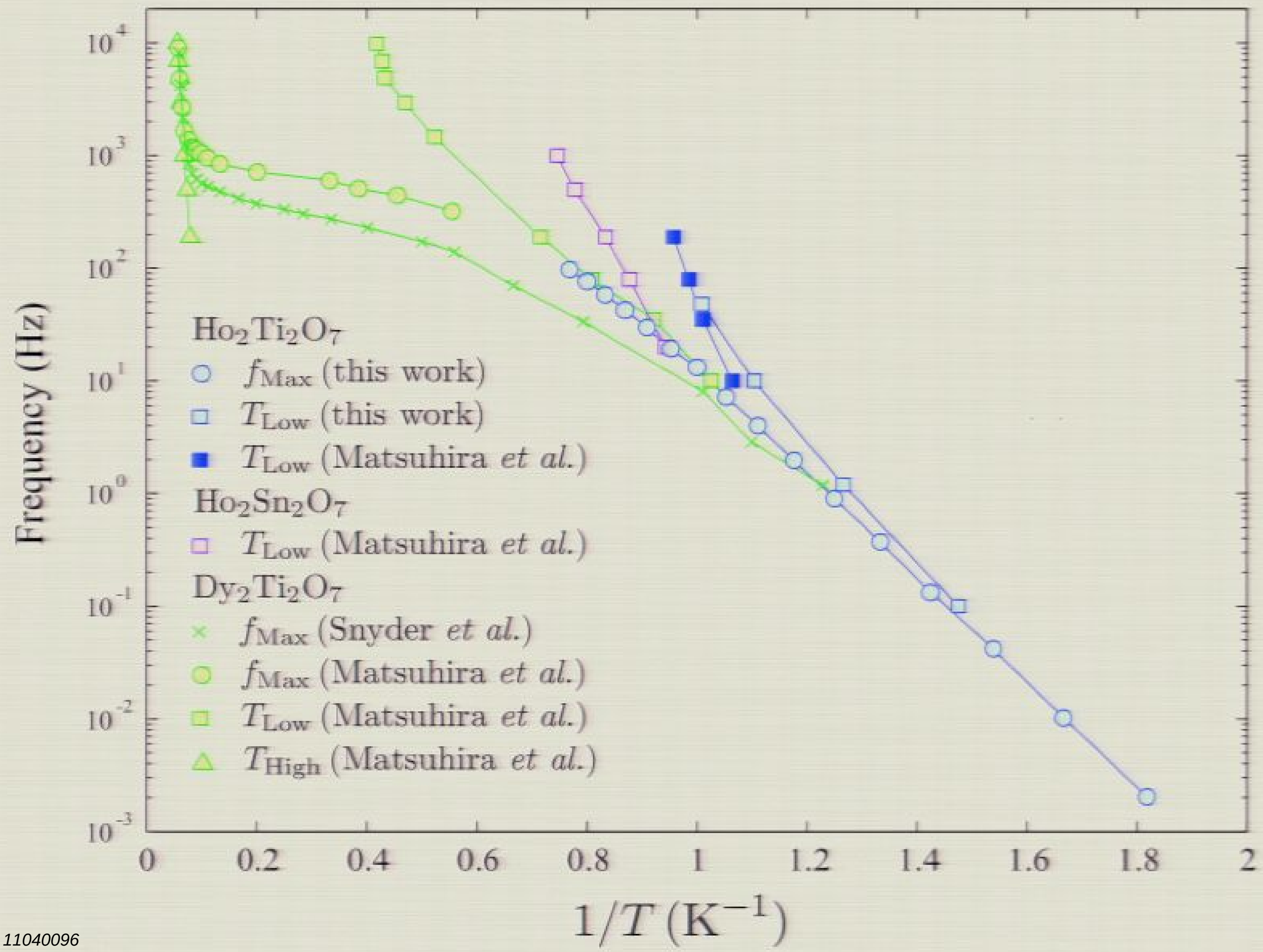


Comparison with other works



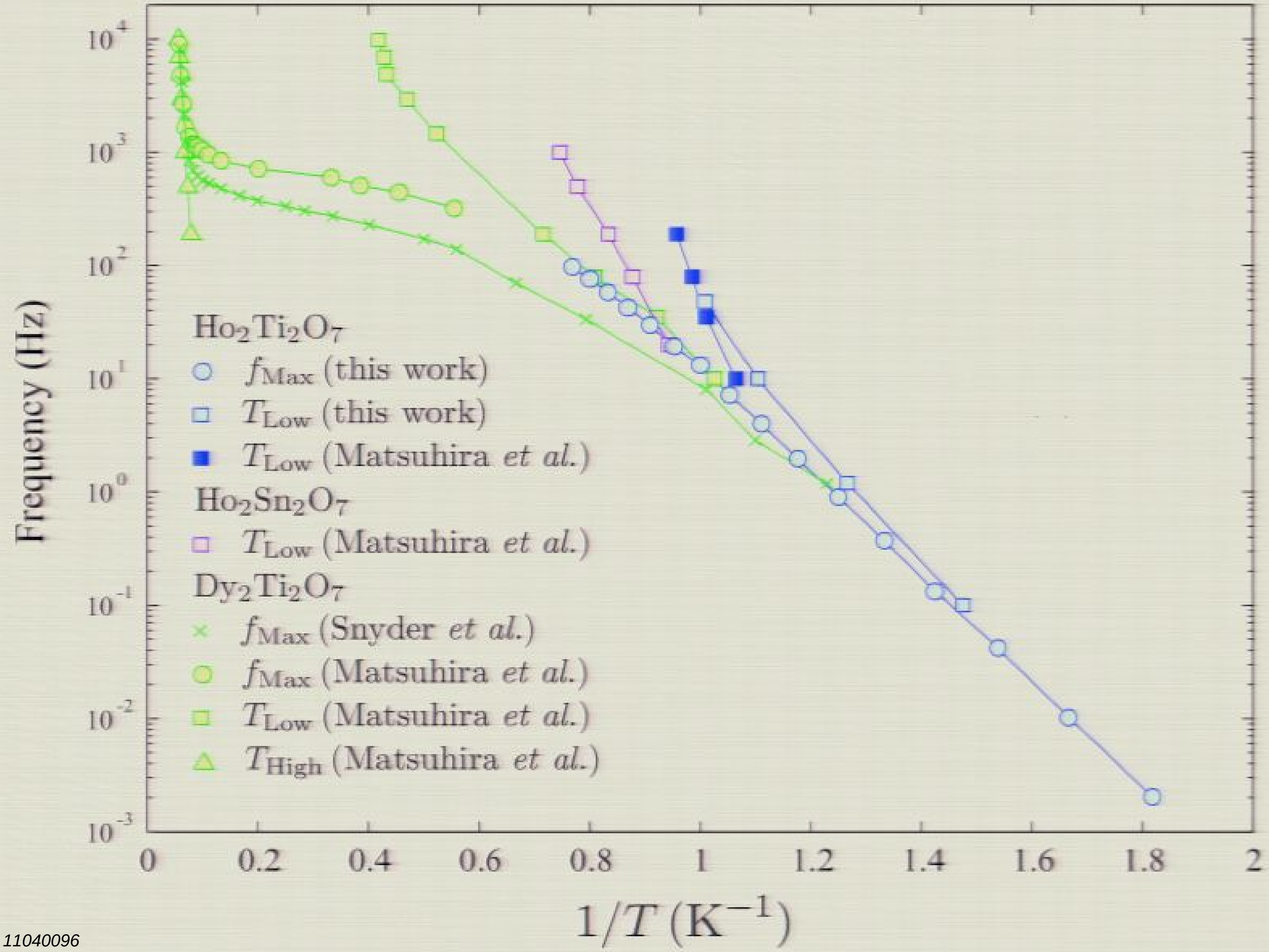
Summary

- Low temperature limit seems follow Arrhenius behaviour with activation energy $6J_{eff} = 10.7K$. Why?

Next:

- Measure DTO to lower temperatures and frequencies, check if this really has $6J_{eff}$ activation energy.
- Work to get accurate measurements at lower and higher frequencies.

Comparison with other works



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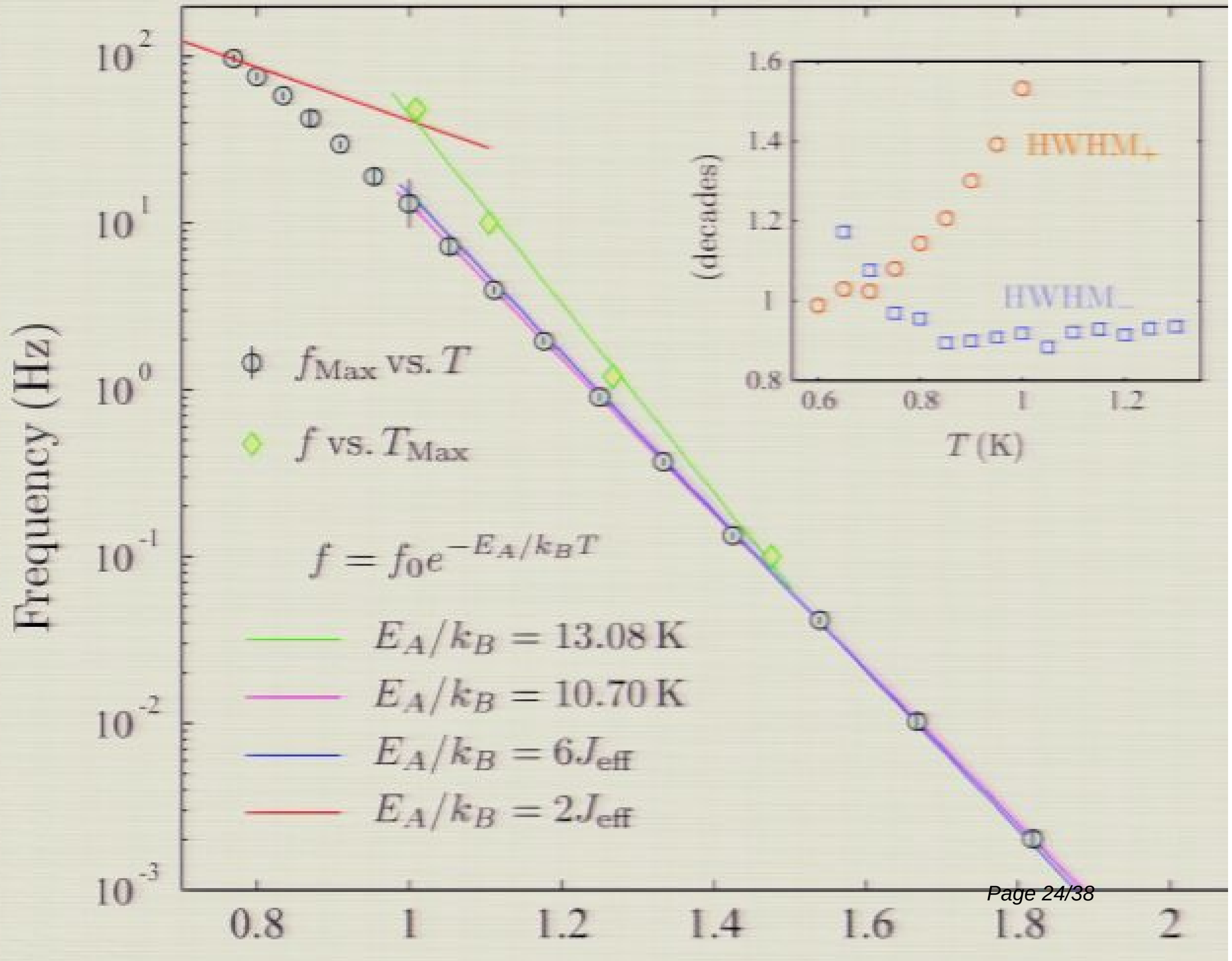
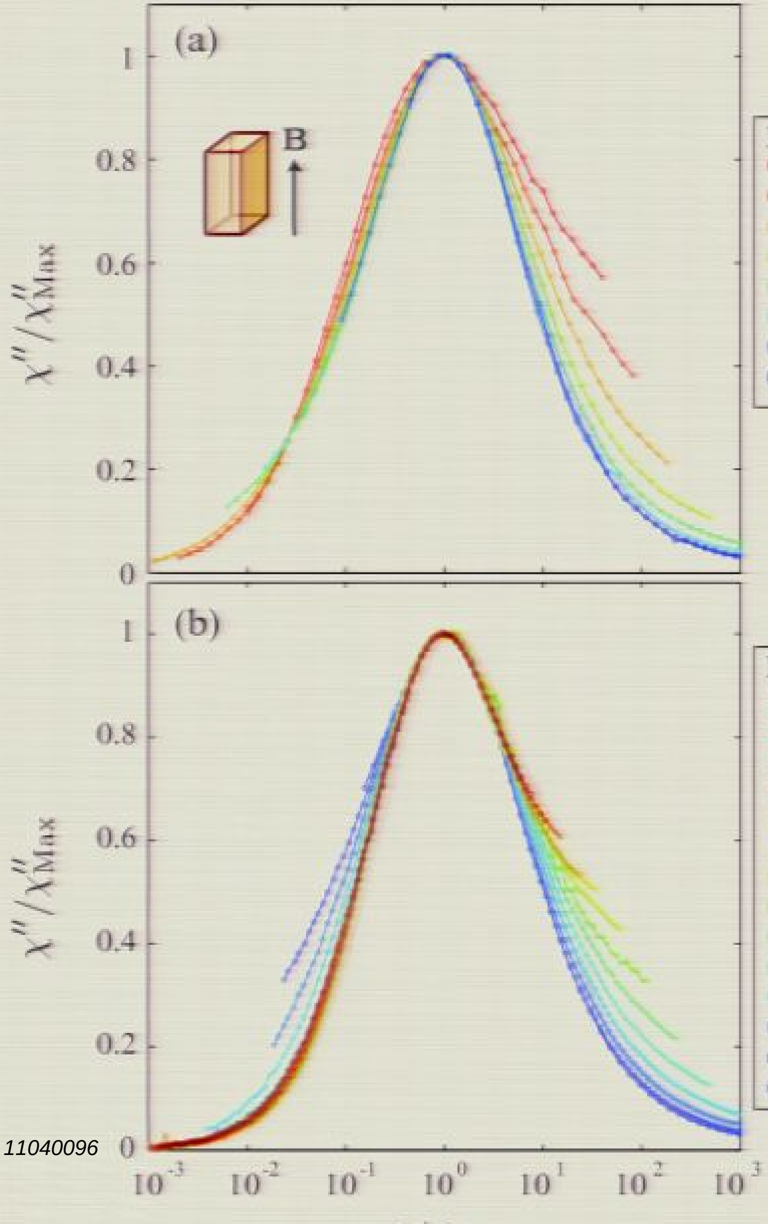
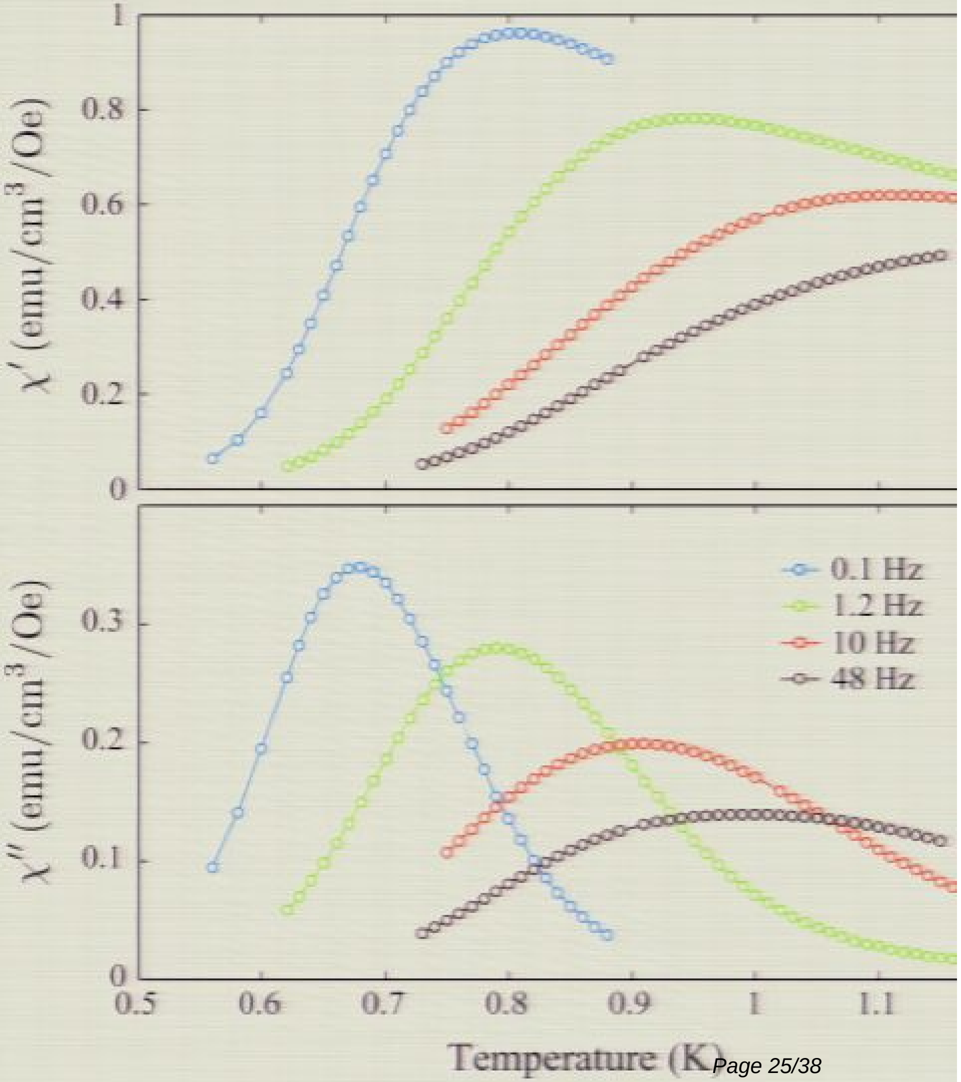


Absorption Spectra and Temperature Scans





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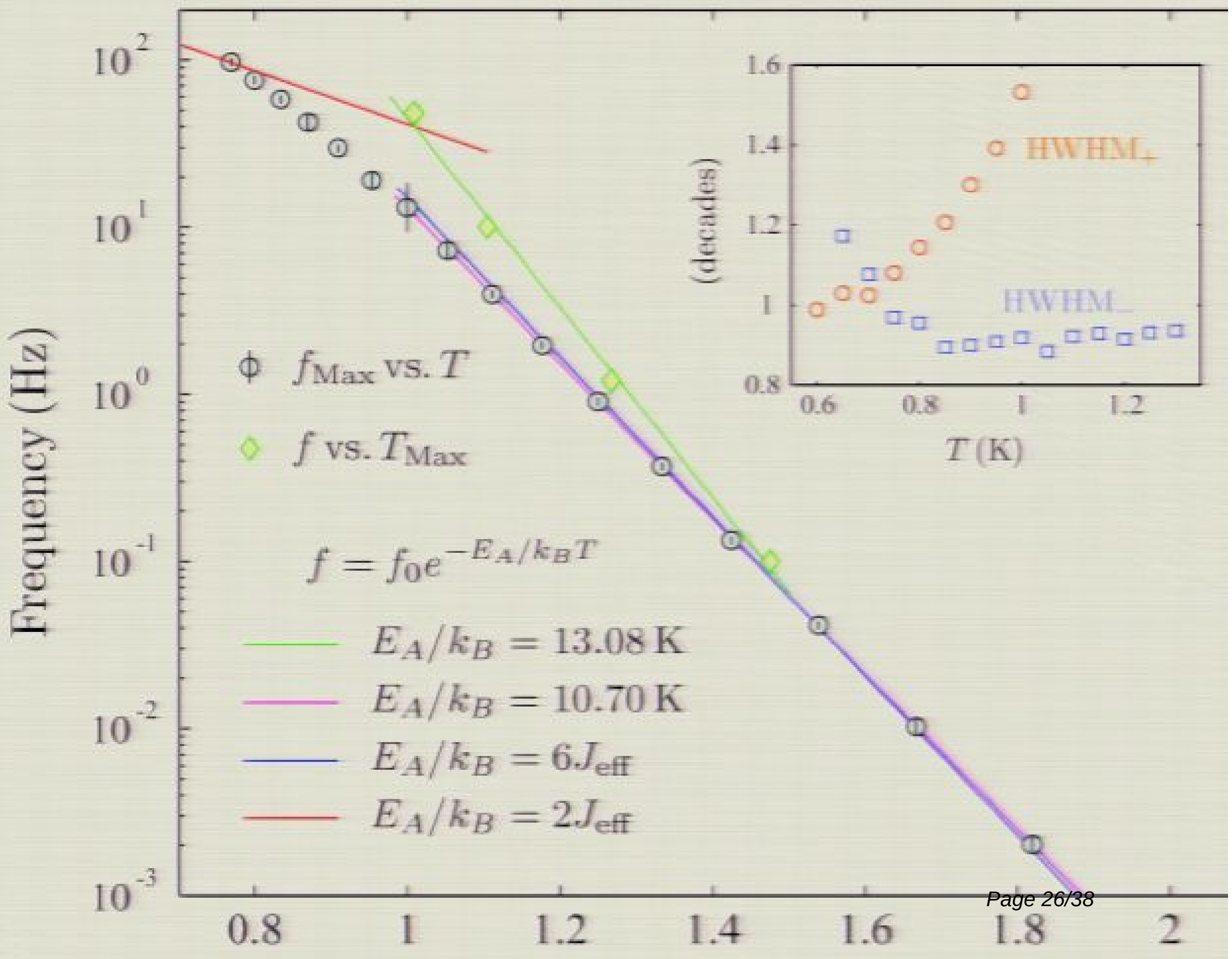

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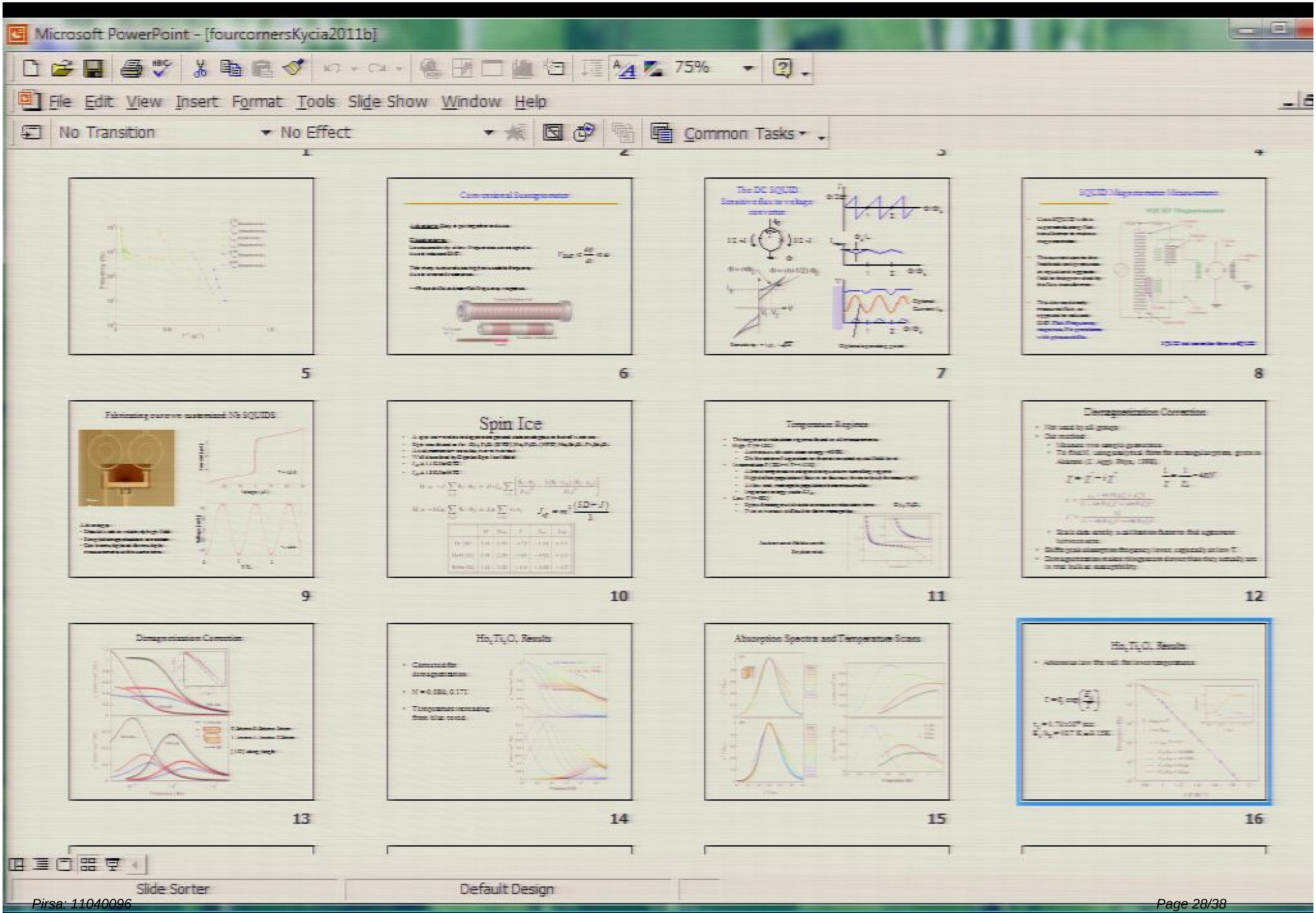
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The DC SQUID:
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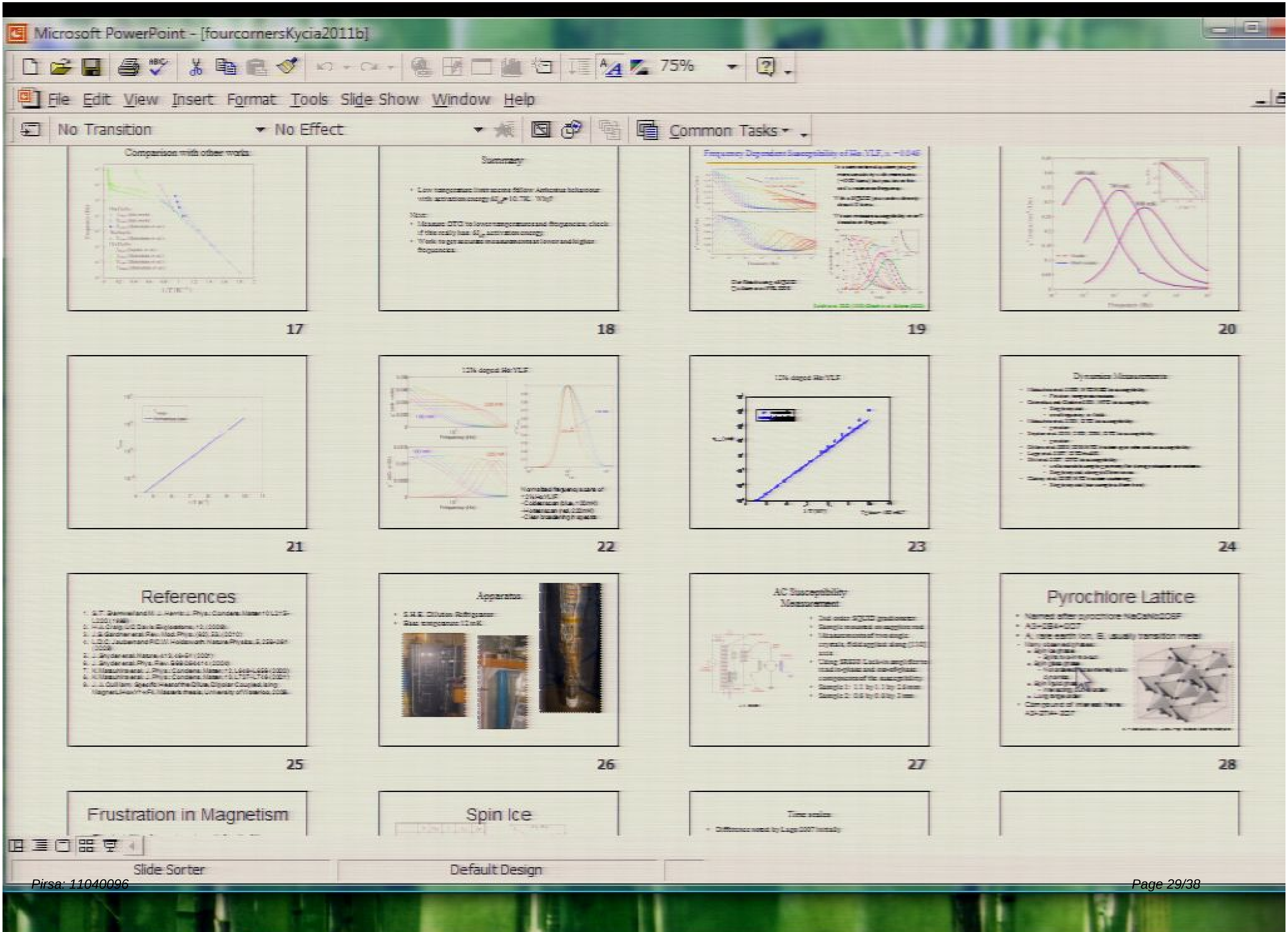

Comparison with other works

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Summary

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12% doped Ho:YLF

22

12% doped Ho:YLF

23

Dynamics Measurements

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Apparatus

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AC Susceptibility Measurement

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Pyrochlore Lattice

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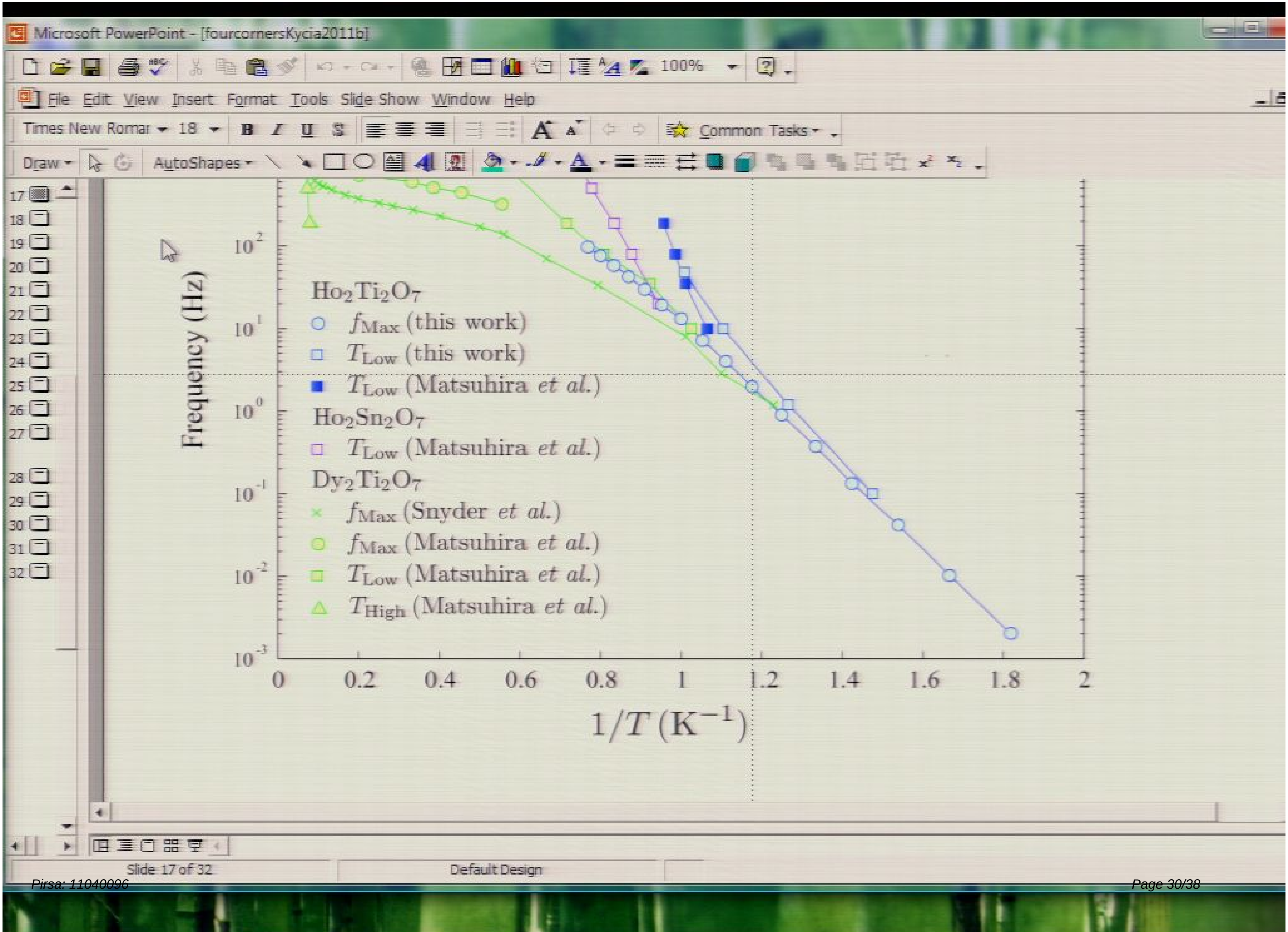
Frustration in Magnetism

Spin Ice

Time scales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Frequency (Hz)
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10^{1}
10^{0}
10^{-1}
10^{-2}
10^{-3}
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0.8
1
1.2
1.4
1.6
1.8
2
$1/T\,(\mathrm{K}^{-1})$
$Ho_2Ti_2O_7$
○ f_{Max} (this work)
□ T_{Low} (this work)
■ T_{Low} (Matsuhira *et al.*)
$Ho_2Sn_2O_7$
□ T_{Low} (Matsuhira *et al.*)
$Dy_2Ti_2O_7$
× f_{Max} (Snyder *et al.*)
○ f_{Max} (Matsuhira *et al.*)
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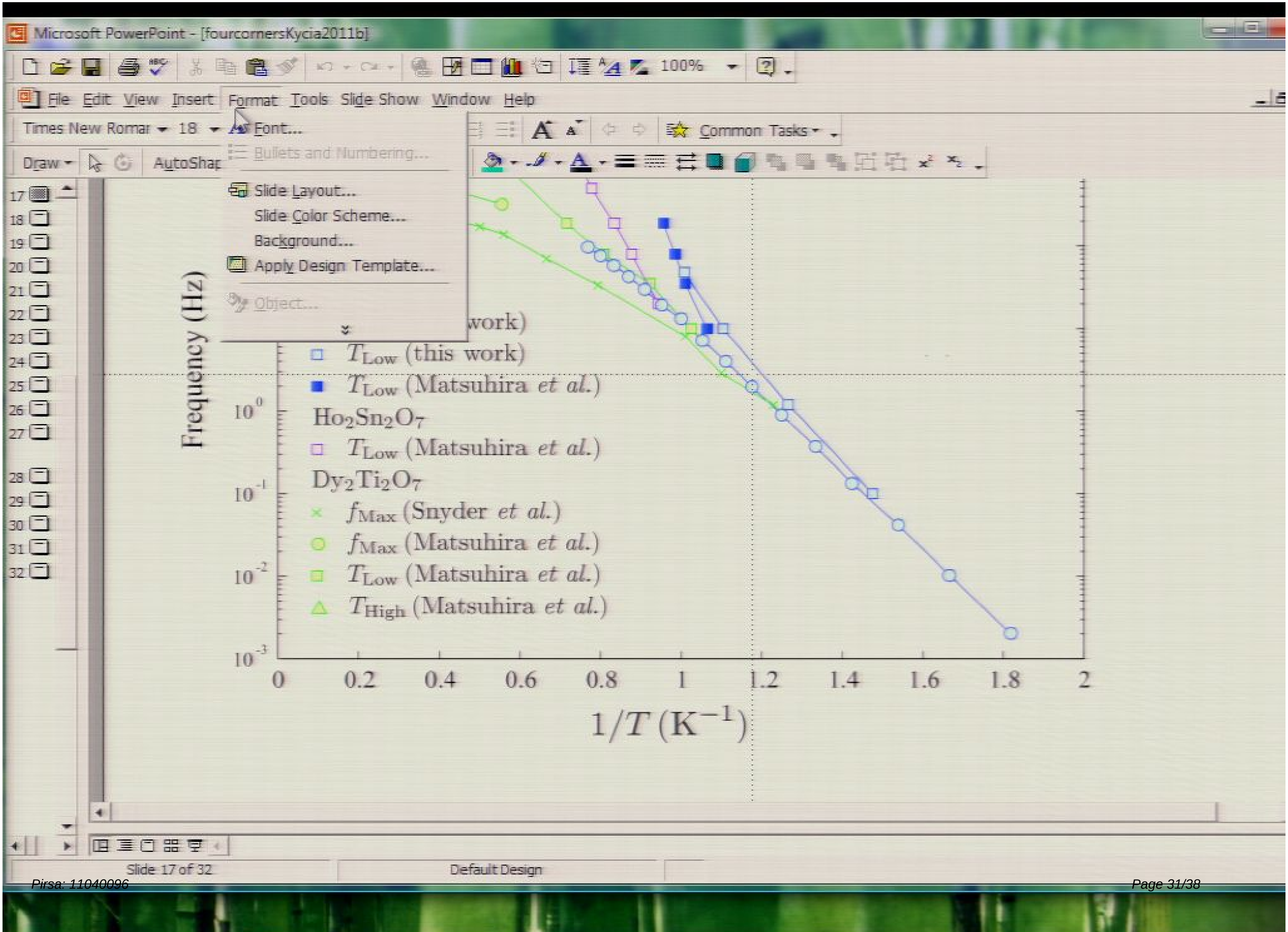
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$Ho_2Sn_2O_7$
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$Dy_2Ti_2O_7$
f_{Max} (Snyder et al.)
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T_{Low} (Matsuhira et al.)
T_{High} (Matsuhira et al.)
Frequency (Hz)
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10^{-2}
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0.2
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$1/T\,(\mathrm{K}^{-1})$
Slide 17 of 32
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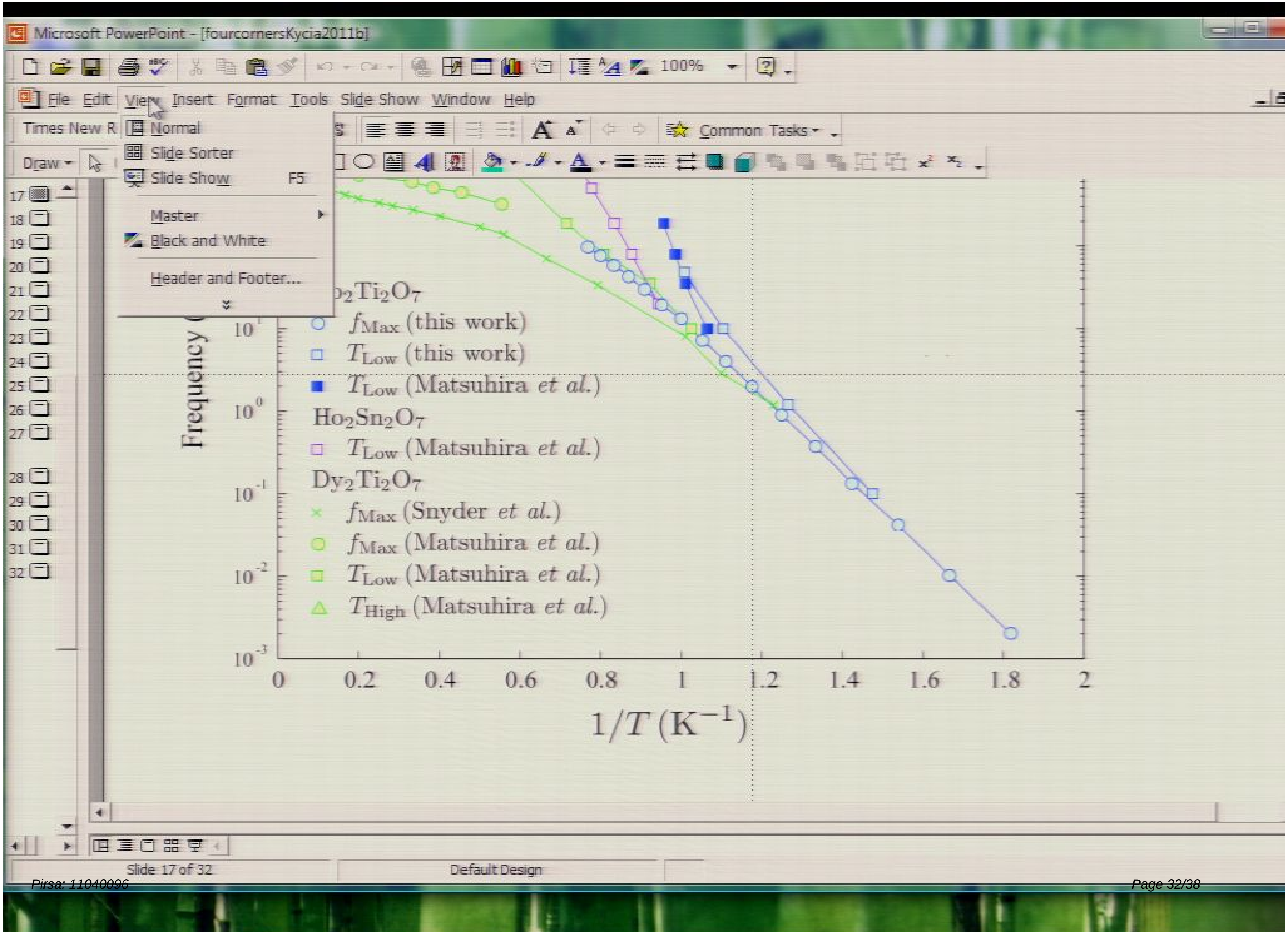
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○ f_{Max} (this work)
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Frequency
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10^0
10^{-1}
10^{-2}
10^{-3}
0
0.2
0.4
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$1/T\,(K^{-1})$
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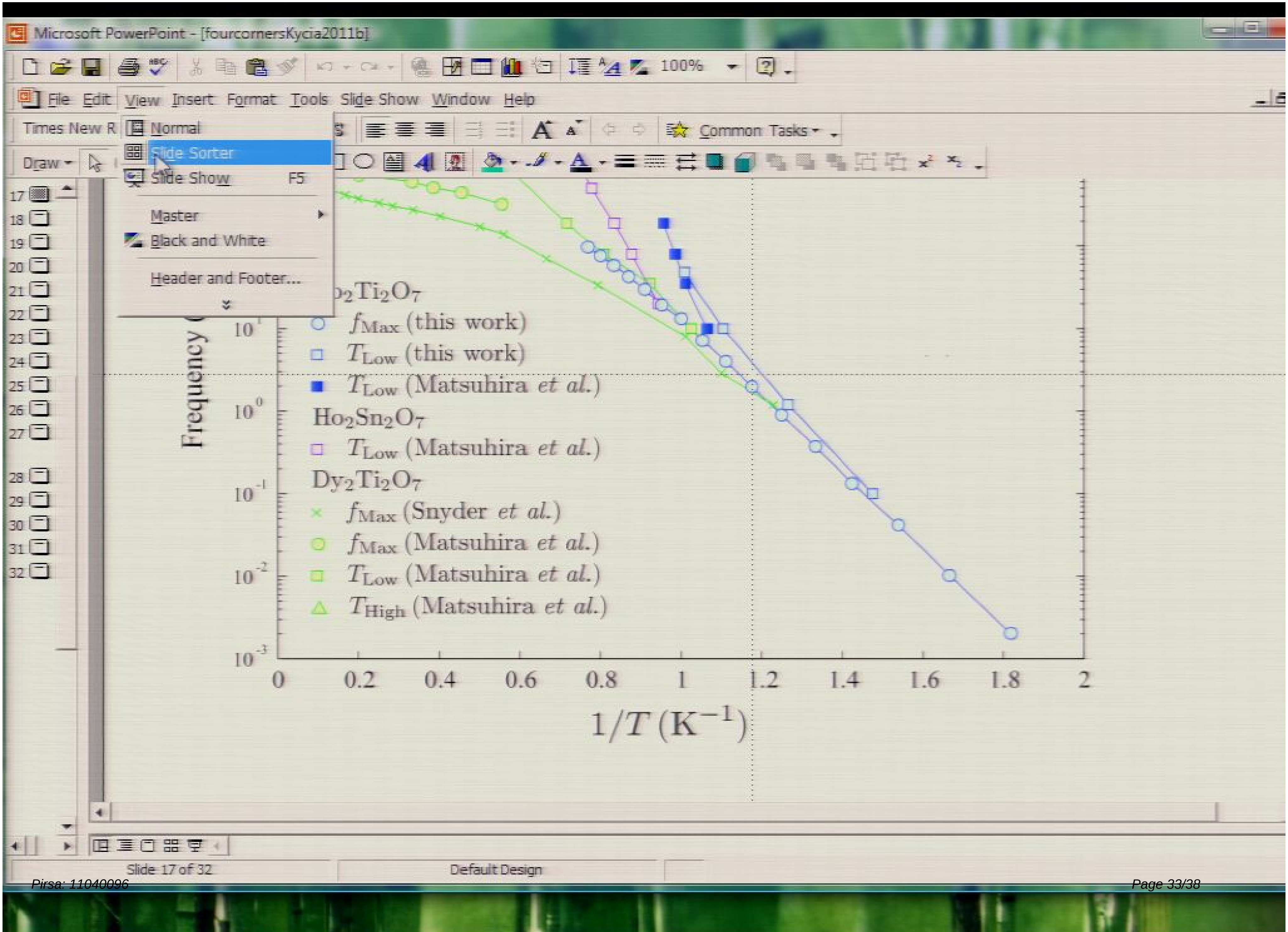
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□ T_{Low} (this work)
■ T_{Low} (Matsuhira *et al.*)
Ho$_2$Sn$_2$O$_7$
□ T_{Low} (Matsuhira *et al.*)
Dy$_2$Ti$_2$O$_7$
× f_{Max} (Snyder *et al.*)
○ f_{Max} (Matsuhira *et al.*)
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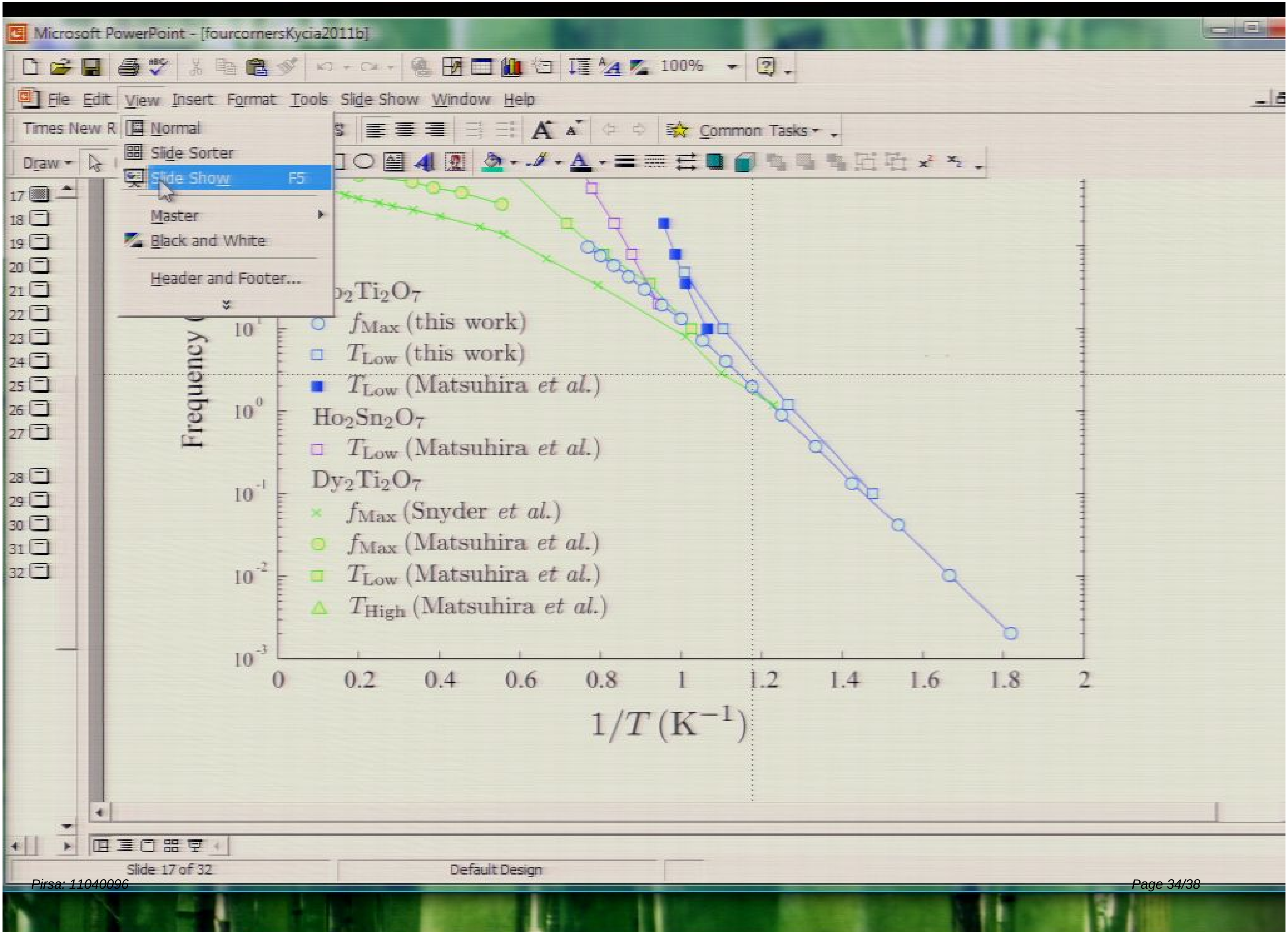
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AC Susceptibility Measurements of $Ho_2Ti_2O_7$

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Spin Ice

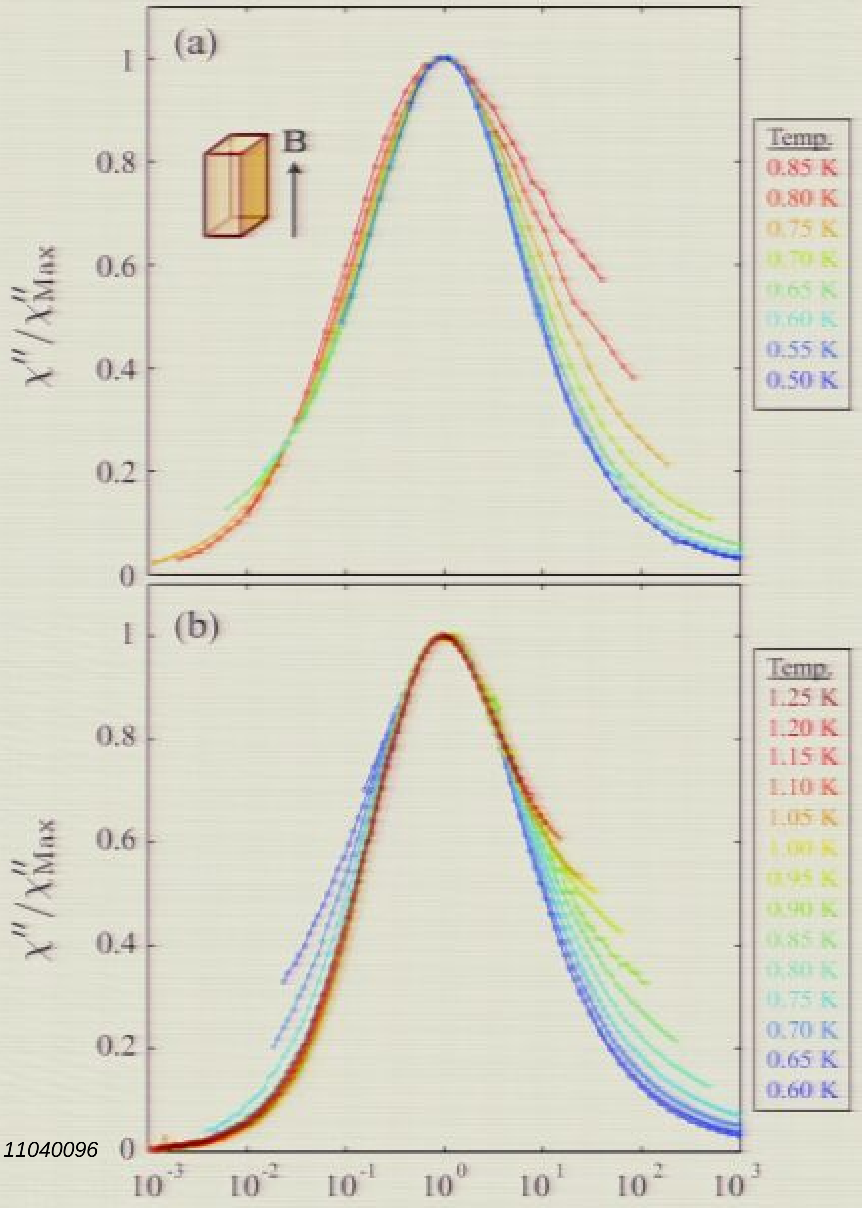
- A spin ice – orders to degenerate ground state analogous to that of water ice
- Spin ices found so far: $Dy_2Ti_2O_7$ (DTO), $Ho_2Ti_2O_7$ (HTO), $Ho_2Sn_2O_7$, $Pr_2Sn_2O_7$
- Axial moments – ice rules, two-in two-out
- Well described by Dipolar Spin Ice Model
- $J_{eff} \cong 1.1K$ for DTO
- $J_{eff} \cong 1.8K$ for HTO

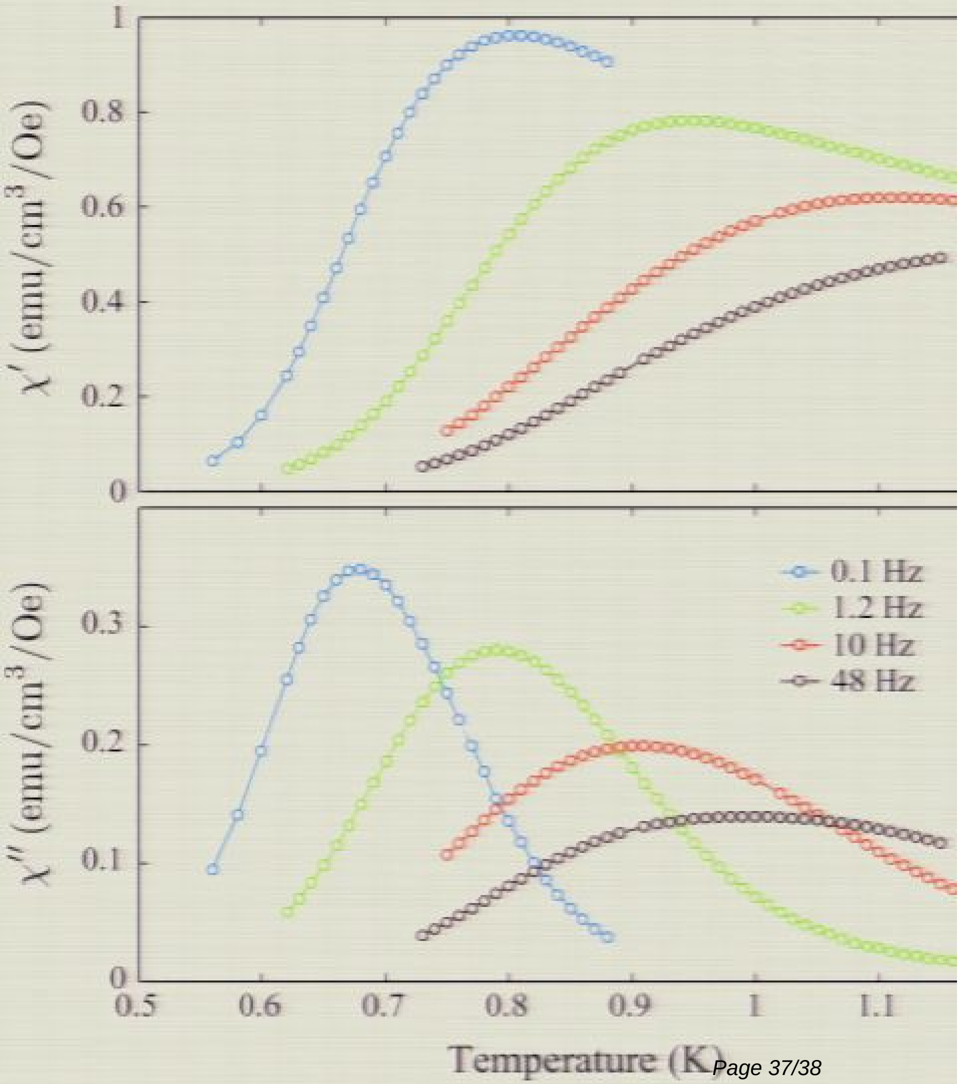
$$\mathcal{H} = -J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + D r_{nn}^3 \sum_{i>j} \left[\frac{\mathbf{S}_i \cdot \mathbf{S}_j}{|\mathbf{r}_{ij}|^3} - \frac{3(\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5} \right]$$

$$\mathcal{H} = -3J_{\text{eff}} \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j = J_{\text{eff}} \sum_{\langle i,j \rangle} \sigma_i \sigma_j \qquad J_{eff} = m^2 \frac{(5D - J)}{3}$$

	D	D_{nn}	J	J_{nn}	J_{eff}
Dy [20]	1.41	2.35	−3.72	−1.24	≈ 1.1
HoTi [21]	1.41	2.35	−1.65	−0.52	≈ 1.8
HoSn [22]	1.41	2.35	−1.0	−0.33	[illegible]

Absorption Spectra and Temperature Scans





Comparison with other works