**Characterization of Nickel Based Spin-One Antiferromagnets**

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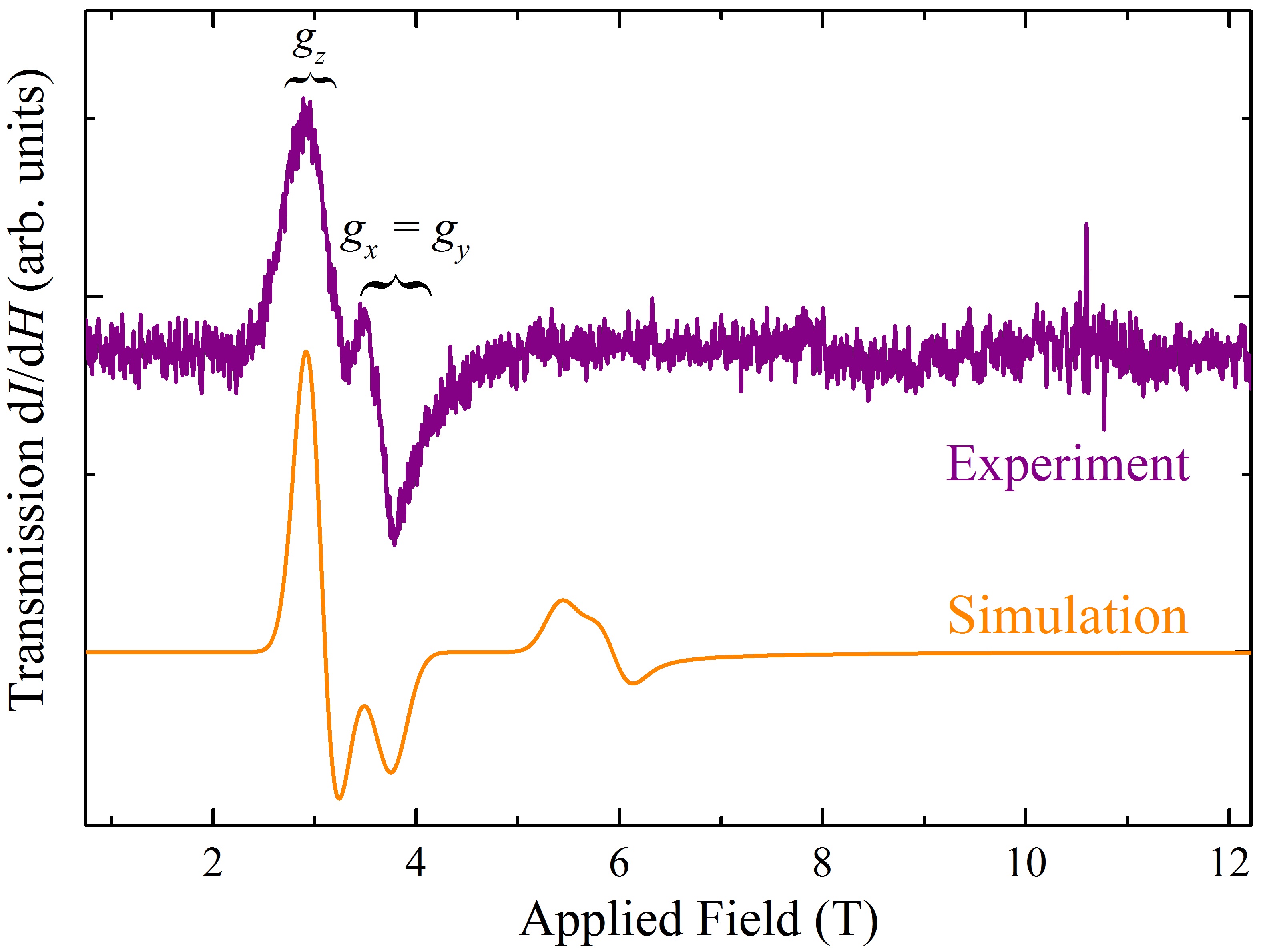
**Introduction**

Much work has been done on low-dimensional Ni2+ *S* = 1 anti-ferromagnetic coordination polymers, but issues still remain in extracting the anisotropy parameters *D* and *E* of compounds that contain magnetic interactions (J) [1,2]. To this end, molecular coordination polymers have been designed such that they contain negligible *J* enabling a much easier determination of *D* and *E* which can then be used to supplement the characterization of structurally similar materials that contain significant magnetic interactions.

**Experiment Details**

High-field, high-frequency ESR spectra at temperatures ranging from 5 to 30 K were recorded on a home-built spectrometer at the EMR facility of the NHMFL, Tallahassee, with the microwave frequencies *ν* = 52-416 GHz. The instrument is a transmission-type device and uses no resonance cavity. The microwaves were generated by a phase-locked Virginia Diodes source, generating frequency of 13 ± 1 GHz, and equipped with a cascade of frequency multipliers to generate higher harmonic frequencies. A superconducting magnet (Oxford Instruments) capable of reaching a field of 17 T was employed.

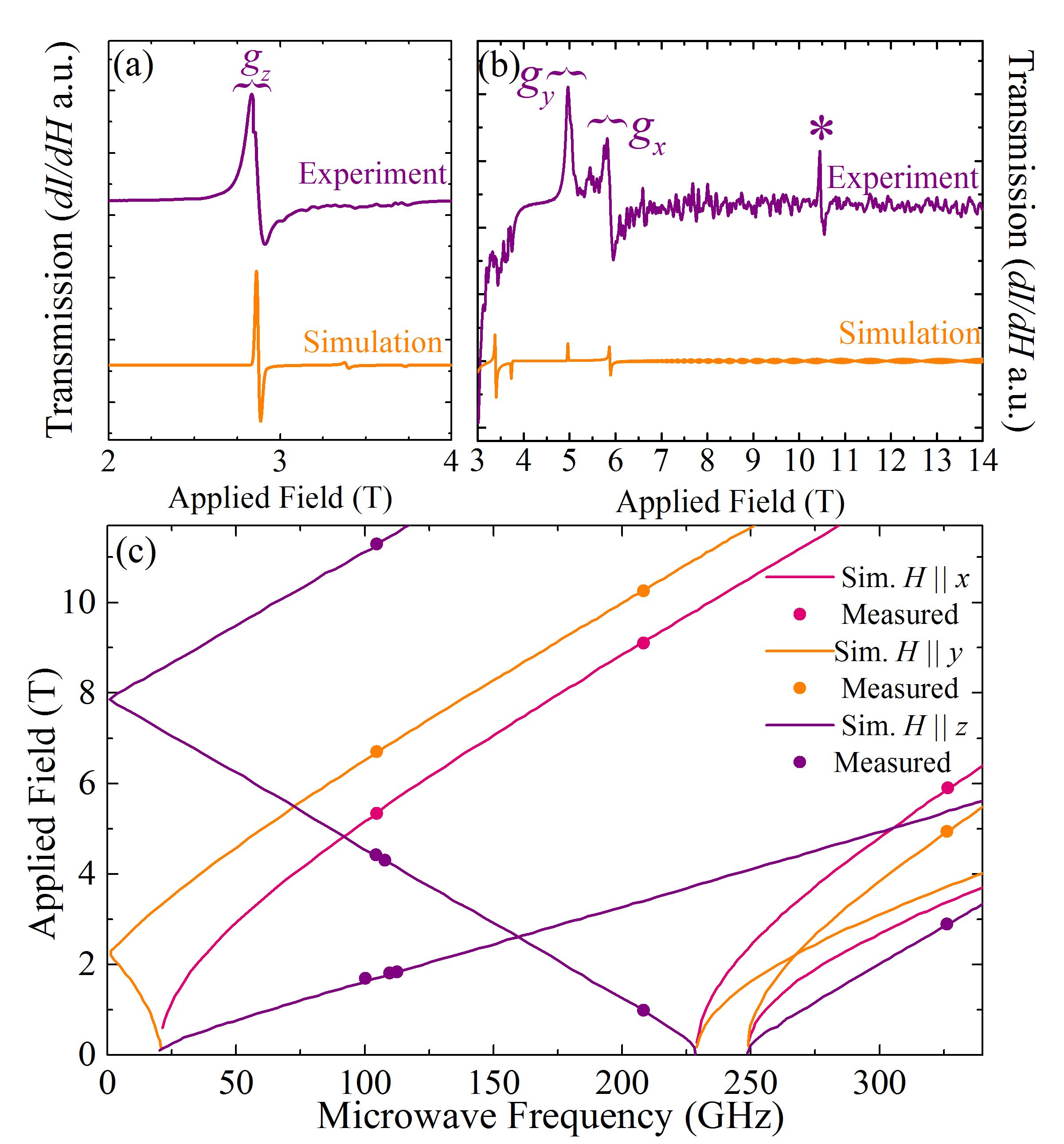
**Results and Discussion**

 Continuous wave powdered ESR measurements (purple) made on the molecular [Ni(H2O)2(3,5-lut)4](BF4)2 are shown in **Fig. 1** along with a simulation (yellow) based on the Hamiltonian:

[1]

with an isotropic *g* = 2.21, *D* = 11.2 K and *E* = 0.27 K. The simulation captures the large resonance. The values of *g*, *D* and *E* are consistent with susceptibility and magnetization measurements. The simulated asymmetric peak centered at 6 T is not present in the measurement. This is believed to be a transition between the two higher energy states. At such low temperatures the lowered occupancy of these levels will correspond to a lowering of the transmission signal.

**Fig. 1** Transmission ESR spectrum vs. applied field for [Ni(H2O)2(3,5-lut)4](BF4)2 at 5 K with a 326.4 GHz source.

Powder transmission ESR measurements (purple) of NiSiF6(4-mepz)4(H2O) are shown in **Fig. 2(a)** and **(b)** [the transmission axis in (b) is expanded 6x that in (a)] along with a simulation (yellow) based on the Hamiltonian in Eq. 1 with values of *gx* = 2.234, *gy* = 2.230, *gz* = 2.160, *D* = 11.44 K and *E* = 0.469 K. The simulation successfully replicates the peaks that correspond to fields at which the separation of the energy levels in Ni2+ ions is equal to *hν*. The asterisk marks a paramagnetic impurity. The position of transitions [**Fig. 2(c)**] were fitted for *S* = 1 ions governed by the Hamiltonian in Eq.1 yielding parameters in excellent agreement with the simulation. These values are consistent with subsequent pulsed-field magnetization measurements that confirm NiSiF6(4-mepz)4(H2O) exhibiting easy-plane anisotropy with *D* = 11.4 K and *E* = 0.5 K.

**Acknowledgements**

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**Fig. 2** (a), (b) Power transmission spectrum vs. applied field for NiSiF6(4-mepz)4(H2O) at 5 K with 326 GHz source and (c) transitions in the ESR spectrum as a function of frequency.

**References**

[1] Liu, J., *et al*., arXiv: 1410.3077, (2015).

[2] Manson, J.L., *et al*., Inorg. Chem., **50**, 5990 (2011).