Supplementary material: Coplanar cavity for strong coupling between photons and magnons in van der Waals antiferromagnet

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I. SCHEMATIC FOR MEASUREMENT CIRCUIT

FIG. S1. (a) Circuit schematic for reflection measurement of NbN SCPW resonators in an Oxford Triton dilution fridge. RF signal from one port of a VNA is carried through a coax line with attenuation at different temperature plates inside the fridge and to the device under test (DUT) through a circulator. Reflected signal from the device is amplified by a Low Noise Factory HEMT amplifier at 4 K plate before sending it back to the other port of the VNA. (b) A lumped element equivalent of the device. Here the parallel combination of L , C and R represent the resonator, C_C represents the coupling capacitor, C_G is a capacitor representing the open-to-ground configuration of the one-port resonator and V_{RF} represents the VNA, which is used for actuation and measurement of the resonator.

Fig. S1 (a) shows schematic of the circuit for reflection measurement of the NbN SCPW resonators. Measurements are done in an Oxford Triton dilution fridge containing a superconducting magnet. Input line of the RF signal includes several attenuators at different stages of the fridge as shown, for proper thermalization of the microwave photons reaching the sample. Input and output paths are separated using a circulator from Quinstar (OXE89 CTH0408KC) before the device. Amplification is done by a Low Noise Factory amplifier (LNF-LNC0.3_14A) attached to the 4 K plate of the fridge. Attained base temperature at sample is 20 mK. The temperature of the sample varies a bit 10-20 mK, so we use the upper range of the temperature. The superconducting coil around the sample is used to apply magnetic field. The measurements are done using an Anritsu (MS46122B) vector network analyzer (VNA). Fig. S1 (b) shows a lumped element equivalent to the device. Note that, because of the signal propagating through the cables, an electrical length dependent phase factor gets multiplied to $S_{11}(f)$ from the device, giving a modified reflectivity $S_{11,mod}(f) = S_{11}(f) \exp(i 2\pi f \frac{L}{v_p})$ where L is the effective length traversed by the signal and v_p is the effective phase velocity of the signal. This electrical length dependent phase part, being a monotonic function of frequency, adds a slope to the background of the phase of the measured signal. Hence, this shows up in the plots of real and imaginary parts of S_{11} as well. By correct-

ing for this overall slope in the background, the pure device response is extracted. We use the functionality available in the VNA to correct for the electrical length while recording data.

II. COMPARISON BETWEEN OVERCOUPLED AND UNDERCOUPLED RESONATORS

FIG. S2. Numeric calculations showing shift in peak height Δh of real part of S_{11} . (a) and (b) show Δh in high Q_i (low k_i) regime for resonator designs with low ($k_e = 10$ MHz) and high ($k_e = 0.1$ MHz) Q_e values respectively (i.e. overcoupled and undercoupled respectively). (c) and (d) show Δh in low Q_i (low k_i) regime for resonators with these two designs respectively. (e) shows a comparison between these two designs of k_e at different k_i . From these plots we observe that an overcoupled design is preferable for good fit in low Q_i regime and an undercoupled design is preferable for good fit in high Q_i regime.

Proper designing of coupling capacitor (C_C) is essential in extracting internal Q of the resonators accurately in regimes with different internal loss rates. Reflection coefficient (S_{11}) of a one-port resonator as a function of frequency is given by

$$S_{11}(f) = 1 - \frac{k_e}{\frac{k_i + k_e}{2} + i(f - f_0)} \quad (\text{S1})$$

where $k_i = \kappa_i/2\pi$ and $k_e = \kappa_e/2\pi$ are internal and external loss rates respectively and $f_0 = \omega_0/2\pi$ is the resonance frequency of the resonator.¹ They are related to internal Q (Q_i) and external

Q (Q_e) by $Q_i = \frac{\omega_0}{\kappa_i} = \frac{f_0}{k_i}$ and $Q_e = \frac{\omega_0}{\kappa_e} = \frac{f_0}{k_e}$. Note that an overall constant phase factor to S_{11} with 0 or π phase can give rise to a dip or peak respectively in the real part of S_{11} , which are equivalent. For understanding effect of chosen Q_e in accuracy of extracted Q_i using Eq. (S1), we do numeric calculations in Mathematica, as shown in Fig. S2. Greater shift in peak height (Δh) of real part of S_{11} for same Q_i variation provides higher accuracy in estimation of Q_i . Fig. S2 (a) and (b) show variation in Δh in high Q_i (i.e. low k_i) regime for high (10 MHz) and low (0.1 MHz) values of chosen k_e respectively. For the higher values of Q_i considered in Fig. S2, these correspond to overcoupled ($Q_i > Q_e$) and undercoupled ($Q_i < Q_e$) designs respectively. High Q_i regime is realized near base temperature of 20 mK and zero magnetic field. Fig. S2 (c) and (d) show variation in Δh in low Q_i (i.e. high k_i) regime for these two choices of k_e . Low Q_i regime is realized as losses are introduced in the system due to increase in temperature and/or magnetic field. Fig. S2 (e) shows a comparison between $k_e = 10$ MHz and $k_e = 0.1$ MHz in terms of variation of Δh with k_i . From these panels we note that in the high Q_i regime undercoupled design (low k_e with $k_e < k_i$) is preferable for good fit, whereas in the low Q_i regime overcoupled design (high k_e with $k_e > k_i$) is preferable for good fit. We use resonators with $Q_e \approx 400$ and $Q_e \approx 22000$ for characterization of the resonators. We find that although lower Q_e is useful for extracting Q_i up to higher magnetic fields it gives high errorbars in the lower magnetic fields. Whereas, using higher Q_e enables us to extract Q_i with higher accuracy in low magnetic field regime, as implied by much lower error bars as shown in main text (Fig.4). This agrees with our comparison in Fig. S2.

III. SIMULTANEOUS FITTING TO REAL AND IMAGINARY PARTS OF S_{11}

We have extracted system parameters by fitting to the real part of S_{11} data using Eq. S1. Simultaneous fitting to both real and imaginary part of S_{11} is also possible for slight improvement. Fig. S3 (a) and (b) show the simultaneous fits to real and imaginary parts of normalized S_{11} obtained from measurement of a resonator with $Q_e \approx Q_i \approx 22000$; this is close to the value obtained by fitting only to the real part of S_{11} as mentioned in main text. Further improvement can be done by implementing ways of continuous background calibration during magnetic field sweep to eliminate the background, which shows slight variation with increasing magnetic field, from the measured signal.

FIG. S3. (a) and (b) Simultaneous fit to real and imaginary parts of S_{11} respectively, of normalized experimental data for an NbN SCPW resonator at zero magnetic field and 20 mK base temperature using Eq. S1.

IV. COMPARISON WITH PREVIOUS REPORTS

TABLE S1. Comparison of Q_i variation with magnetic field with previous reports

Material	Resonator geometry	Flux trapping scheme	Max B_{para} (for $Q_i > 10^3$)	Max B_{perp} (for $Q_i > 10^3$)
Nb [2]	SCPW	No	2.7 T (loaded $Q > 10^3$)	24 mT (loaded $Q > 2.5 \times 10^4$)
NbTiN [3]	SCPW	Yes	6 T ($Q_i > 10^5$)	45 mT (20 mT for $Q_i > 10^5$)
NbTiN nanowire [4]	Nanowire	No	6 T ($Q_i > 2 \times 10^5$)	400 mT ($Q_i > 10^4$)
NbN (our work)	SCPW	No	1 T	100 mT

Table S1 shows a comparison of highest applied magnetic field, in orientations parallel and perpendicular to the SCPW plane, up to which $Q_i > 10^3$ is retained, with previous reports for other type-II superconductors. We observe that even in absence of any substrate surface treatment and flux trapping schemes, our NbN resonators retain $Q_i > 10^3$ up to parallel fields comparable to and perpendicular field twice the maximum value reported previously for an SCPW resonator. Further implementation of substrate surface treatments and flux trapping schemes are expected to increase this maximum field of high Q_i retention even more.

FIG. S4. Characterization of NbN SCPW resonators as a function of microwave power and temperature. (a) and (b) Variation of Q_i with increasing number of microwave photons and temperature, respectively, for intrinsic silicon substrate [Inset of (b): Variation of $\Delta f_0/f_{0,0}$ with temperature for the same]. Note that the error-bars are of size comparable to the markers. (c) and (d) Same for resonators on a sapphire substrate.

V. CHARACTERIZATION OF RESONATORS WITH MICROWAVE POWER AND TEMPERATURE

For characterizing the fabricated resonators with respect to experimental parameters, we study the internal loss of the resonators by varying experimental conditions such as drive power and temperature. Dependence of resonance characteristics on P_{RF} is performed to investigate the performance of these resonators at and below single photon power level. The microwave power dependence of Q_i is shown in Fig. S4 (a) for intrinsic silicon substrate and in Fig. S4 (c) for sapphire substrate. From Fig. S4 (a) and (c) we observe a small increasing trend in Q_i as the mean number of microwave photons in the cavity is increased. Such a behavior suggests the presence of two-level systems (TLS).⁵ At higher power, TLS get saturated and result in higher Q_i . For all the magnetic field dependent measurements discussed in the manuscript, we use a measurement

power equivalent to approximately 10^3 photons in the resonator. Temperature dependence of the Q_i has been shown in Fig. S4 (b) for intrinsic silicon substrate and in Fig. S4 (d) for sapphire substrate (the insets show temperature dependence of $\frac{\Delta f_0}{f_{0,0}}$). As a general trend, above $T \approx 4$ K f_0 of the resonators show a downward shift. This reduction in f_0 can be attributed to the increasing kinetic inductance due to reduction in the number density of available Cooper pairs. Furthermore, for the same reason, increase in number density of quasiparticles causes internal losses to increase, thereby lowering Q_i . A downward shift in the relative frequency shift below $T \approx 1.5$ K confirms the presence of TLS.⁵

VI. CHARACTERIZATION OF RESONATORS ON SAPPHIRE SUBSTRATE WITH MAGNETIC FIELD

FIG. S5. Characterization of an NbN SCPW resonator on sapphire substrate (NbN film thickness 94 nm) as a function of magnetic field. (a) and (b) Variation of Q_i and $\Delta f_0/f_{0,0}$ for parallel and perpendicular field orientations respectively.

Fig. S5 (a) and (b) show variation of Q_i and $\Delta f_0/f_{0,0}$ with magnetic field parallel (B_{para}) and perpendicular (B_{perp}) to the SCPW plane respectively. This dependence is similar to that of resonators on intrinsic silicon substrate as shown in main text.

FIG. S6. Relative shift in resonance frequency $\Delta f_0/f_{0,0}$ of NbN resonators on intrinsic silicon substrate. (a) and (b) Variation in $\Delta f_0/f_{0,0}$ for the overcoupled resonators with NbN film thicknesses 217 nm and 72 nm in parallel and perpendicular magnetic field respectively. (c) and (d) Variation in $\Delta f_0/f_{0,0}$ for the undercoupled resonators with NbN film thickness 80 nm in parallel and perpendicular magnetic field respectively.

VII. VARIATION IN $\Delta f_0/f_{0,0}$ WITH MAGNETIC FIELD FOR RESONATORS ON INTRINSIC SILICON SUBSTRATE

Fig. S6 (a) and (b) show the variation of $\Delta f_0/f_{0,0}$ for resonators on intrinsic silicon substrate with film thicknesses 217 nm and 72 nm with magnetic field parallel and perpendicular to the SCPW plane respectively. These show the nearly quadratic and linear dispersion of resonance frequency in parallel and perpendicular magnetic field respectively. The slope of the linear dispersion has been used to calculate electronic diffusion constant D . Fig. S6 (c) and (d) show the same for resonators with undercoupled design.

FIG. S7. Hysteresis in NbN SCPWs with magnetic field. (a) and (b) Hysteresis in an NbN SCPW resonator on intrinsic silicon substrate with respect to direction of magnetic field sweep, with field parallel and perpendicular to the SCPW respectively.

VIII. HYSTERESIS WITH MAGNETIC FIELD

We observe some amount of hysteresis in the NbN SCPW resonators depending upon the direction of magnetic field sweep. Fig. S7 (a) and (b) show hysteresis in Q_i of a resonator on intrinsic silicon substrate. We observe that for the perpendicular field, Q_i is higher in down-sweep of field compared to the up-sweep; whereas, for the parallel field, we observe regimes with higher as well as lower Q_i in down-sweep compared to up-sweep. The perpendicular field case is similar to previous report and is similar to prediction from Norris-Brandt-Indenbom (NBI) model with inhomogeneous current density.⁶ But the presence of two regimes in case of parallel field sweep suggests the presence of grain boundaries^{7,8} in the NbN film, which is also apparent from the SEM image of an NbN film as shown in the inset of Fig.1 in the main text.

IX. LINEAR FIT FOR FINDING D

The electronic diffusion constant (D) is given by $D = \frac{8k}{\pi e} \frac{\beta^2 k_B T_C}{\beta^2 - f_0^2 L_g}$ as described in main text, where k is the negative of slope of $\frac{\Delta f_0}{f_{0,0}}$ vs B_{perp} plot. Fig. S8 shows a linear fit to the $\frac{\Delta f_0}{f_{0,0}}$ vs B_{perp} data for an NbN SCPW resonator on intrinsic silicon substrate. The estimated electronic diffusion constant is $D = 5.01 \times 10^{-4} \text{ m}^2 \text{ s}^{-1}$.

FIG. S8. Linear fit to the $\frac{\Delta f_0}{f_{0,0}}$ vs B_{perp} data for determination of electronic diffusion constant (D) for an NbN SCPW resonator on intrinsic silicon substrate.

X. TRANSMISSION SPECTROSCOPY OF CrCl_3

Chromium trichloride (CrCl_3) is a van der Waals antiferromagnet with weak interlayer exchange. Fig. S9 (a) shows the crystal structure of CrCl_3 . The localized magnetic moments in CrCl_3 are provided by the Cr^{3+} ions shown as black spheres. In any atomic layer of CrCl_3 , the magnetic moments are in plane, shown by blue arrows, and are coupled by ferromagnetic exchange. Magnetic moments in consecutive planes are antiferromagnetically coupled. Due to weak interlayer exchange of $1.6 \mu\text{eV}$, antiferromagnetic resonance (AFMR) is observed at GHz frequencies in CrCl_3 whereas, most antiferromagnets have resonance modes in THz frequency range. This allows realization of coupling of the AFMR modes in CrCl_3 with cavity modes in coplanar waveguide resonators operating in the low GHz frequencies.

To study the transmission spectra of CrCl_3 , the crystal flake was placed on a copper CPW transmission line PCB and secured with Apezion grease to protect it from ambient. The measurement was done in a He flow cryostat. As shown in Fig. S9 (b), using a vector network analyzer, microwave signal was sent from port-1 to the transmission line and the transmitted signal was amplified before being received at the port-2 of the VNA. DC magnetic field was applied parallel to the plane of the PCB and perpendicular to the segment of the transmission line containing the CrCl_3 crystal (i.e. perpendicular to the RF current at the position of the CrCl_3). The transmission spectra was obtained by measuring $S_{21}(f)$ at each value of DC magnetic field as it was swept up starting from zero field. The derivative of the transmission spectra with respect to the DC magnetic field was then plotted against the magnetic field and frequency, as shown in Fig. 4(a) of the manuscript. Using the derivative divide method,⁹ the background signal can be removed from the

FIG. S9. (a) Crystal structure of CrCl₃ shows van der Waals stacking of antiferromagnetically oriented consecutive atomic layers (black spheres denote the Cr³⁺ ions and blue arrows depict associated moments). (b) Measurement schematic for the transmission spectroscopy of a CrCl₃ crystal flake placed on a transmission line made of copper on a PCB. (c) Schematic showing a CrCl₃ crystal flake placed on the central part of an NbN SCPW resonator for the magnon-photon coupling measurement. (d) Optical microscopy image of the crystal in the part shown by a rectangle in (c).

transmission spectra without calibration of the microwave circuits. This data can then be fit to an anti-Lorentzian to determine the linewidths of the modes. A detailed study of the different modes seen in CrCl₃ is presented in ref [10] and [11] .

Fig. S9 (c) shows the schematic of CrCl₃ crystal flake placed on an NbN SCPW resonator and Fig. S9 (d) shows the optical microscopy image of the grease covered CrCl₃ crystal on the NbN resonator at the current antinode of the resonator (the part shown by a rectangle in Fig. S9 (c)). As the crystal covers the entire NbN resonator, the effective volume of interaction between the microwave photons in the NbN resonator and the magnons in CrCl₃ is determined mainly by the mode volume of the magnetic component of the RF field of the resonator.

XI. FIT TO MAGNON DISPERSION

FIG. S10. (a) Polynomial fit (solid lines) to magnetic field dispersions of bare acoustic and optical AFMR modes for CrCl_3 crystal flake placed on copper transmission line. (b) Non-linear model fit to mode dispersion corresponding to magnon-photon coupling data of Fig.4(b) of the manuscript using magnetic field dependent functional form of the eigenmodes of three mode coupling Hamiltonian matrix (the dots represent the experimental data and the solid lines represent the fit).

The reflection coefficient (S_{11}) for the system with two non-interacting magnon modes coupled with a cavity photon mode can be written according to input-output theory^{12–14} as

$$S_{11}(f) = 1 - \frac{k_e}{i(f - f_0) + \frac{k_i + k_e}{2} + \frac{|g_1/(2\pi)|^2}{i(f - f_1) + \frac{\gamma_1/(2\pi)}{2}} + \frac{|g_2/(2\pi)|^2}{i(f - f_2) + \frac{\gamma_2/(2\pi)}{2}}} \quad (\text{S2})$$

From Eq.S2 we can obtain the functional form for $S_{11}(B, f)$ in case of CrCl_3 placed at current antinode of an NbN resonator, by inserting the dispersion relation of each mode with magnetic field. As the B field for this measurement is parallel to the plane of the SCPW resonator, the dispersion of cavity resonance frequency (f_0) can be taken to be constant within the field range of interest for observing magnon-photon coupling. We take this to be $f_0 = 4.6$ GHz. For k_i and k_e , we use the corresponding zero field values for the cavity (at zero field, cavity and AFMR modes are off-resonance), $k_i = \frac{\kappa_i}{2\pi} = 0.0064$ GHz and $k_e = \frac{\kappa_e}{2\pi} = 0.0145$ GHz. For the field dependent functional form of the acoustic (f_1) and optical (f_2) AFMR mode frequencies in CrCl_3 , we fitted linear and quadratic polynomials respectively to their field dispersions obtained from the transmission spectroscopy measurement using copper transmission line. Also, for the linewidths of the AFMR modes, we used $\frac{\gamma_1}{2\pi} = 0.33$ GHz and $\frac{\gamma_2}{2\pi} = 0.42$ GHz, as their variation with magnetic field is small.¹¹ This fitting is shown in Fig. S10(a). For estimation of the coupling strengths, we used a

three mode coupling Hamiltonian matrix, whose elements are described in the manuscript. Using magnetic field dependent functional form of the eigenvalues of this matrix, we did a non-linear model fitting to the mode dispersions of Fig.4(b) of manuscript. This fitting gives $\frac{g_1}{2\pi} = 0.57$ GHz and $\frac{g_2}{2\pi} = 0.37$ GHz. The fitting has been shown in Fig. S10(b). From Fig.4(b) of the manuscript, we note that there is a slight downward shift in cavity resonance frequency with increasing field, in the range 0 - 0.038 T which could possibly be attributed to change in magnetic state of CrCl_3 as it undergoes spin-flop transition.^{15–17} Beyond this transition, initially antiparallel spins get in canted orientation with higher fields and give rise to net magnetic moment. We fit the data between 0.038 T and 0.380 T. The cooperativity has been calculated using these values as mentioned in the manuscript. We note that all the features of the observed data are not captured by the model considered. For further understanding some aspects of the data, a microscopic modeling and more detailed analysis is required.

As the crystal covers the entire SCPW resonator, the effective volume of interaction between the magnons and the microwave photons is essentially determined by the mode volume of the magnetic component of the RF field associated with the resonator. This mode volume can be calculated using the formula $V_{\text{SCPW}} = \frac{E_{\text{SCPW}}}{\frac{1}{2\mu_0}B_{\text{max}}^2}$, where E_{SCPW} is the energy stored in the magnetic field of the SCPW resonator and B_{max} is the maximum RF magnetic field of the SCPW resonator. We performed COMSOL simulation of the resonator to find E_{SCPW} and B_{max} , which gives a magnetic mode volume approximately 0.006 mm^3 for the SCPW resonator. Furthermore, using the lattice parameters¹⁶ of CrCl_3 and the magnetic component of the vacuum RF field of the SCPW resonator^{18,19}, we calculate the magnon-photon coupling strength per spin to be $g_0 = 7.93$ Hz and corresponding net coupling strength for the mode volume considered to be $g = g_0\sqrt{N} = 1.09$ GHz, where N is the total number of spins in the considered volume. Better understanding of the difference between $\frac{g}{2\pi}$ that are experimentally obtained, 0.57 GHz and 0.37 GHz for the acoustic and the optical modes respectively, and the estimate of 1.09 GHz, requires microscopic magnetic modeling. Furthermore, the coupling strengths associated with the acoustic and the optical magnons depend on exact geometry and symmetry, and hence a more accurate calculation taking into account symmetry and microscopic details is required for proper estimation for the coupling strengths.

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