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Coherent pumping of high-momentum magnons by light

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We propose and model a method to excite a large number of coherent magnons with high momentum in optical cavities. This is achieved by two counterpropagating optical modes that are detuned by the frequency of a selected magnon, similar to stimulated Raman scattering. In submillimeter-size yttrium iron garnet spheres, a milliwatt laser input power generates 10^6 – 10^8 coherent magnons. The large magnon population enhances Brillouin light scattering, a probe suitable to access their quantum properties.

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Magnets are crucial for fast, nonvolatile, and robust data storage as well as candidate materials for logic devices and interconnects [1]. Magnetic insulators, such as yttrium iron garnet (YIG) [2], are interesting since they can transport information over long distances via spin waves quantized into magnons [3,4], without the Ohmic dissipation of spin transport in metals. The magnons couple to microwaves [5–7], electric currents [1,3,8], mechanical motion [9–12], and light [13,14]. The high crystal quality of YIG promises long coherence times [6,7], opening prospects for “quantum magnonics” [15], the field that strives to employ magnons to store, process, and transfer information in a quantum coherent manner. Photons can become a coherent interface to manipulate and probe these magnons.

The gigahertz magnons in ferro(ferri)magnets interact with light by inelastic (Brillouin) light scattering (BLS) [13]. By selecting the wave vector of the input and output photons, e.g., by an optical cavity, specific magnon modes can be excited [16]. The interaction can be large enough [17,18] to cool [19] or herald (generating single magnon states) [20] them, making BLS a promising probe into their quantum nature. Present experiments focus on the long-wavelength “Walker” (including the “Kittel”) magnons in optical resonators [21–25]. These have a small overlap with the light fields and corresponding low intrinsic scattering efficiency, but become observable because a large magnon density can be resonantly excited by microwaves. On the other hand, magnons with wavelengths ~ 100 – 500 nm in the dipolar-exchange regime have almost perfect overlap with the photon modes in magnetic spheres [18], but couple only very weakly to microwaves (as do the relevant magnons in magnetic vortices [17]).

Here, we present a theory showing that optical lasers can pump a large number ($\sim 10^6$ – 10^8) of high-momentum magnons similar to the resonant excitation of Kittel magnons by microwaves. We exploit the torques exerted by light on the magnetization by the inverse Faraday and Cotton-Mouton effects [26], which are proportional to the intensity of the

electric field component [26] or, more precisely, the product of the photon numbers at the incident and scattered frequencies. Exposing the sample to two phase-coherent lasers that differ in frequency by a magnon excitation strongly enhances Brillouin scattering [27]. Here we develop the theory of stimulated light scattering by magnons in optical resonators such as sketched in Fig. 1. Two counterpropagating lasers feed whispering gallery modes (WGMs) of a YIG sphere via a proximity coupler such as a fiber or a prism [21–23,28]. The WGMs are separated spectrally by ~ 1 – 10 GHz, which can be easily tuned into resonance with a magnon by an applied magnetic field. The two populated WGMs form a spatially periodic torque field that excites magnons with matching wavelength. While we focus here on spherical magnets, the formalism is valid for any magneto-optical cavity, including planar [29,30] and cylindrical [17] geometries.

We consider a minimal model of two WGM modes $\{W_r, W_b\}$ resonantly interacting with a single magnon mode M (see Fig. 1). We first formulate heuristic rate equations for the magnon number, $n_m^{(sc)}$ (“sc” stands for semiclassical), followed by a more rigorous quantum Langevin treatment. In the steady state, the energy balance of the processes in Fig. 1 leads to the photon number in the blue sideband W_b with frequency ω_b [31],

$$N_b = \frac{4K_b}{(\kappa_b + K_b)^2} \frac{P_b}{\hbar\omega_b}, \quad (1)$$

which is governed by the input light power P_b , the decay rate κ_b in the isolated sphere, and the leakage rate K_b into the proximity coupler. An analogous expression holds for the photon number N_r in the red sideband W_r . Since optomagnonic couplings are small, we disregarded the backaction exerted by magnons on photons. The reaction rate for anti-Stokes scattering $W_r + M \rightarrow W_b$ is $R_b = R_b^{(0)} n_m^{(sc)} N_r (N_b + 1)$, while for the reverse (Stokes) scattering $R_r = R_r^{(0)} (n_m^{(sc)} + 1) (N_r + 1) N_b$. According to the Fermi’s golden rule, $R_{b,r}^{(0)} = 2\pi |g|^2 \Lambda_{b,r}(\Delta)$, where g is the matrix element of the Hamiltonian between initial and final states (see below), the detuning

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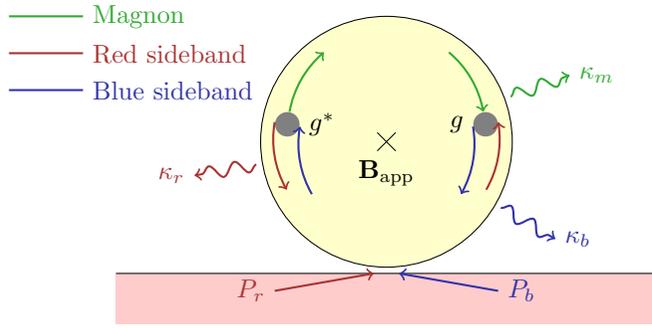


FIG. 1. A (massive) sphere of a magnetic insulator, such as YIG, with a proximity optical coupler, such as a fiber or a prism. Two oppositely propagating laser beams excite two whispering gallery modes with decay rates $\kappa_{r,b}$. The photon-magnon scattering coherently amplifies the magnon amplitude competing with the thermalization rate κ_m .

$\Delta \equiv \omega_b - \omega_r - \omega_m$, and

$$\Lambda_{b,r} = \frac{1}{2\pi} \frac{(\kappa_{b,r} + K_{b,r})}{\Delta^2 + (\kappa_{b,r} + K_{b,r})^2/4}, \quad (2)$$

with $(\kappa_{b,r} + K_{b,r})^{-1}$ as the photon's lifetime.

Magnons are lost at a rate $R_{\text{eq}} = \kappa_m(n_m^{(\text{sc})} - n_{\text{eq}})$ where κ_m^{-1} is the magnon lifetime and n_{eq} is the equilibrium (Planck) distribution

$$n_{\text{eq}} = \left[\exp\left(\frac{\hbar\omega_m}{k_B T}\right) - 1 \right]^{-1}. \quad (3)$$

In the steady state $R_b + R_{\text{eq}} = R_r$,

$$n_m^{(\text{sc})} = \frac{R_r^{(0)} N_b (N_r + 1) + \kappa_m n_{\text{eq}}}{\kappa_m + R_b^{(0)} N_r (N_b + 1) - R_r^{(0)} (N_r + 1) N_b}. \quad (4)$$

Equation (4) agrees with the more rigorous result below only when $R_b^{(0)} = R_r^{(0)}$, because here we ignored the correlation between the forward and backward reactions. Furthermore, the above treatment does not distinguish between coherent and thermal magnons. For sufficiently large $N_{r,b}$, $n_m^{(\text{sc})}$ may diverge which is an artifact of ignoring magnon nonlinearities, but such large drives are unrealistic (shown below).

We consider the three-particle Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}_{\text{om}}$ with noninteracting part

$$\hat{H}_0 = \hbar\omega_r \hat{a}_r^\dagger \hat{a}_r + \hbar\omega_b \hat{a}_b^\dagger \hat{a}_b + \hbar\omega_m \hat{m}^\dagger \hat{m}, \quad (5)$$

where $\{\hat{a}_r, \hat{a}_b, \hat{m}\}$ are the annihilation operators for $\{W_r, W_b, M\}$, respectively. To leading order in the magnon operators the optomagnonic Hamiltonian is [16]

$$\hat{H}_{\text{om}} = \hbar g \hat{a}_r \hat{a}_b^\dagger \hat{m} + \hbar g^* \hat{a}_r^\dagger \hat{a}_b \hat{m}^\dagger. \quad (6)$$

In the Heisenberg picture, the statistical averages $\langle \hat{X}(t) \rangle = \text{Tr}[\hat{X}(t)\hat{\rho}]$, where the density matrix $\hat{\rho} = \hat{\rho}_0$ is a direct product of an arbitrary state of the sphere (magnons and WGMs) and a coherent photon state of the laser input.

The equation of motion for the blue sideband envelope operator $\hat{W}_b \triangleq \hat{a}_b e^{i\omega_b t}$ reads [19,20,32]

$$\frac{d\hat{W}_b}{dt} = -ig\hat{W}_r\hat{M}e^{i\Delta t} - \frac{\kappa_b + K_b}{2}\hat{W}_b - \sqrt{\kappa_b}\hat{b}_b + \sqrt{K_b}\hat{A}_b, \quad (7)$$

where the first term on the right-hand side is the optomagnonic scattering generated by the commutator $[\hat{a}_b, \hat{H}]$ in the Heisenberg equation. The second term is the decay of photons inside the sphere, $\propto \kappa_b$, and into the coupler, $\propto K_b$. \hat{b}_b is the annihilation field operator of a bath mode that interacts with W_b satisfying the commutation relations $[\hat{b}_b(t), \hat{b}_b^\dagger(t')] = \delta(t - t')$ and averages $\langle \hat{b}_b(t) \rangle = \langle \hat{b}_b^\dagger(t') \hat{b}_b(t) \rangle = 0$. Without input $K_b = 0$ and optomagnonic coupling $g = 0$, the steady state of Eq. (7) is the thermal equilibrium state [33] with no photons since $k_B T \ll \hbar\omega_b$. The input field operator \hat{A}_b of the propagating photons in the coupler [19,20,32] satisfies the commutation relations $[\hat{A}_b(t), \hat{A}_b^\dagger(t')] = \delta(t - t')$, with average

$$\langle \hat{A}_b(t) \rangle = \sqrt{\frac{P_b}{\hbar\omega_b}} e^{i\mathcal{W}_b(t)}, \quad (8)$$

and correlator

$$\langle \hat{A}_b^\dagger(t') \hat{A}_b(t) \rangle = \frac{P_b}{\hbar\omega_b} e^{i[\mathcal{W}_b(t) - \mathcal{W}_b(t')]} \quad (9)$$

The photons suffer from phase noise that we model by a classical random walk $\mathcal{W}_b(t) = \sqrt{\kappa_{\text{ph}}} \int_0^t \mathcal{N}(x) dx$, with dephasing rate $\sqrt{\kappa_{\text{ph}}} \cdot \kappa_{\text{ph}} / (2\pi)$ typically ranges from hertz to megahertz [34], much smaller than the typical inverse lifetimes in a resonator $\kappa_{\text{ph}} \ll \kappa_b \sim 2\pi \times (0.1-1)$ GHz. The phase noise is taken to be white with $\langle \mathcal{N} \rangle_{\text{cl}} = 0$ and $\langle \mathcal{N}(t)\mathcal{N}(t') \rangle_{\text{cl}} = \delta(t - t')$.

Since Eq. (7) is linear, $\hat{W}_b(t) = \hat{W}_{b,\text{opt}}(t) + \hat{W}_{b,\text{om}}(t)$, with optical contribution at large times $t \gg 1/\kappa_b$ being

$$\hat{W}_{b,\text{opt}}(t) = - \int_0^t e^{-(\kappa_b + K_b)(t-\tau)/2} [\sqrt{\kappa_b} \hat{b}_b(\tau) + \sqrt{K_b} \hat{A}_b(\tau)] d\tau \quad (10)$$

includes the thermal noise and input from the coupler. In the steady state and for $\kappa_{\text{ph}} \ll \kappa_b$, we get the commutation relations

$$[\hat{W}_{b,\text{opt}}(t), \hat{W}_{b,\text{opt}}^\dagger(t')] = e^{-(\kappa_b + K_b)|t-t'|/2}, \quad (11)$$

the average

$$\langle \hat{W}_{b,\text{opt}}(t) \rangle = \sqrt{N_b} e^{i\mathcal{W}_b(t)}, \quad (12)$$

and correlator

$$\langle \hat{W}_{b,\text{opt}}^\dagger(t') \hat{W}_{b,\text{opt}}(t) \rangle = N_b e^{i[\mathcal{W}_b(t) - \mathcal{W}_b(t')]} \quad (13)$$

with N_b from Eq. (1). The optomagnonic scattering $W_r + M \rightarrow W_b$ contributes

$$\hat{W}_{b,\text{om}}(t) = -ig \int_0^t e^{-(\kappa_b + K_b)(t-\tau)/2} \hat{W}_r(\tau) \hat{M}(\tau) e^{i\Delta\tau} d\tau. \quad (14)$$

For the red sideband $\hat{W}_r(t) = \hat{W}_{r,\text{opt}}(t) + \hat{W}_{r,\text{om}}(t)$, with $\hat{W}_{r,\text{opt}}(t)$ analogous to Eq. (10) and scattering contribution

$$\hat{W}_{r,\text{om}}(t) = -ig^* \int_0^t e^{-(\kappa_r + K_r)(t-\tau)/2} \hat{W}_b(\tau) \hat{M}^\dagger(\tau) e^{-i\Delta\tau} d\tau. \quad (15)$$

The magnon envelope operator $\hat{M}(t) \triangleq \hat{m}(t) e^{i\omega_m t}$ obeys

$$\frac{d\hat{M}}{dt} = -ig^* \hat{W}_r^\dagger \hat{W}_b e^{-i\Delta t} - \frac{\kappa_m}{2} \hat{M} - \sqrt{\kappa_m} \hat{b}_m, \quad (16)$$

where the stochastic magnetic field, $\hat{b}_m(t)$, is generated by magnon-phonon [35], magnon-magnon [36,37], surface roughness [38], and (rare earth) impurity scattering [39–42]. When $k_B T/\hbar \gg \kappa_m$, which for $\kappa_m \sim 2\pi \times 1$ MHz [21–23] means $T \gg 50$ μ K, we can write $\langle \hat{b}_m(t) \rangle = 0$, $\langle \hat{b}_m^\dagger(t') \hat{b}_m(t) \rangle = n_{\text{eq}} \delta(t - t')$, and $\langle \hat{b}_m(t') \hat{b}_m^\dagger(t) \rangle = (n_{\text{eq}} + 1) \delta(t - t')$, with average magnon number n_{eq} [see Eq. (3)]. When $g = 0$, the steady state of Eq. (16) is the Planck distribution of the magnon number at temperature T [33], given in Eq. (3).

The optical torque $\propto g^*$ in Eq. (16) generates coherent magnons. To leading order in $g/\kappa_{r,b}$,

$$\frac{d\langle \hat{M} \rangle}{dt} = -i\bar{\omega} \langle \hat{M} \rangle - ig^* \sqrt{N_r N_b} e^{-i\Delta t + i\mathcal{W}(t)} - \frac{\kappa_{\text{eff}}}{2} \langle \hat{M} \rangle, \quad (17)$$

where

$$\bar{\omega} = |g|^2 \Delta \left(\frac{4N_b}{4\Delta^2 + (\kappa_r + K_r)^2} - \frac{4N_r}{4\Delta^2 + (\kappa_b + K_b)^2} \right) \quad (18)$$

is a shift in the magnon frequency, $\mathcal{W} = \mathcal{W}_b - \mathcal{W}_r$ is the phase noise with variance $2\kappa_{\text{ph}}$, and the effective damping $\kappa_{\text{eff}} = \kappa_m + \bar{\kappa}_b - \bar{\kappa}_r$. Here

$$\bar{\kappa}_b = \frac{4|g|^2 N_r (\kappa_b + K_b)}{4\Delta^2 + (\kappa_b + K_b)^2} \quad (19)$$

is proportional to the reaction rate of $W_r + M \rightarrow W_b$ [see Eq. (2)] and $\bar{\kappa}_r$ is given by $r \leftrightarrow b$. Equation (17) leads to the steady state

$$\lim_{t \rightarrow \infty} \langle \hat{M}(t) \rangle = \frac{-ig^* \sqrt{N_r N_b}}{i(\bar{\omega} - \Delta) + \kappa_{\text{ph}} + \kappa_{\text{eff}}/2} e^{-i\Delta t + i\mathcal{W}(t)}, \quad (20)$$

where we assumed ergodicity of \mathcal{W} . The phase noise of the input laser fields is imprinted on the magnon amplitude.

We estimate the magnitude of the effects for an input laser with typical vacuum wavelength ~ 1 μ m and $\omega_r \approx \omega_b \approx \omega_{\text{opt}} = 2\pi \times 300$ THz. For a YIG sphere, the optical quality can be as high as $\omega_r/\kappa_r = \omega_b/\kappa_b = 10^6$ [12] and is limited by light absorption (for frequencies at which the magneto-optical coupling is significant). The magnon linewidth $\kappa_m = 2\pi \times 1$ MHz and we adopt the optomagnonic coupling $|g| = 2\pi \times 200$ Hz [18] for a sphere of radius $R = 300$ μ m (with $|g| \propto 1/R$). We assume low phase noise $\kappa_{\text{ph}} \ll \kappa_m$ which can otherwise be absorbed into κ_{eff} [cf. Eq. (20)]. An external magnetic field can tune ω_m into resonance at $\Delta = 0$. For impedance-matched optical coupling $\kappa_{r,b} = K_{r,b} = \kappa_{\text{opt}}$, the total magnetic damping

$$\kappa_{\text{eff}} = \kappa_m \left(1 + \frac{P_r - P_b}{P_{\text{sat}}} \right) \quad (21)$$

with saturation power (to be interpreted below)

$$P_{\text{sat}} = \frac{\hbar \kappa_m \omega_{\text{opt}} \kappa_{\text{opt}}^2}{2|g|^2} = 1 \text{ W}. \quad (22)$$

For moderate $P_{r,b} \sim 1$ – 10 mW, $\kappa_{\text{eff}} \approx \kappa_m$ is limited by the intrinsic (Gilbert) damping of the magnet. For the large coupling $|g| = 2\pi \times 4$ kHz predicted for a magnetic vortex in a thin magnetic disk [17], $P_{\text{sat}} = 3.5$ mW.

Our main result is the number of coherently excited magnons

$$n_c = \lim_{t \rightarrow \infty} |\langle \hat{M}(t) \rangle|^2 = \frac{P_r P_b}{P_{\text{crit}}^2}, \quad (23)$$

in terms of the critical power

$$P_{\text{crit}} = \frac{\hbar \kappa_{\text{eff}} \omega_{\text{opt}} \kappa_{\text{opt}}}{2|g|}, \quad (24)$$

which is a measure for the input power required to generate significant coherent dynamics. It is smaller than P_{sat} by a factor $\kappa_{\text{opt}}/|g| \sim 10^6$. With $\kappa_{\text{eff}} \approx \kappa_m$, $P_{\text{crit}} = 1$ μ W is in experimental reach. We predict a large $n_c = 10^6$ – 10^8 for $P_{r,b} \sim 1$ – 10 mW. In a magnetic vortex [17], $P_{\text{crit}} = 50$ nW and $n_c = 5 \times (10^8$ – $10^{10})$. A typical exchange-dipolar surface magnon has a volume $V_{\text{mag}} \sim 10^{-3} V_{\text{sph}}$ [18] where V_{sph} is the volume of the sphere. $n_c = 10^6$ and $R = 300$ μ m thus correspond to an average density 8 μm^{-3} , while the peak density is an order of magnitude higher. Using $g \propto 1/R$ [18] the density has a strong scaling $\propto R^{-5}$.

Next we demonstrate that the coherence of the excited magnons is very high (in the absence of absorption heating by the lasers), i.e., the fluctuations around the coherent component $\delta \hat{M} = \hat{M} - \langle \hat{M} \rangle$ are very small, by solving Eq. (16). We employ a weak-coupling approximation [19] by expanding up to the leading terms in $\hat{W}_{x,\text{om}}$. When $\delta \hat{M}$ varies much slower than $\kappa_{r,b}$ (shown *a posteriori* to be equivalent to high optical damping $\kappa_{r,b} \gg \kappa_{\text{eff}}$) we can replace $\delta \hat{M}(\tau) \rightarrow \delta \hat{M}(t)$ in the expression of photons Eqs. (14) and (15). Furthermore, we ignore correlations between photons and magnons beyond second order in g , which is equivalent to replacing photon operators by their mean-field average (see [19] for intermediate steps). Then Eq. (16) reduces to

$$\frac{d}{dt} \delta \hat{M} = - \left(i\bar{\omega} + \frac{\kappa_{\text{eff}}}{2} \right) \delta \hat{M} - \sqrt{\kappa_{\text{eff}}} \hat{b}_{\text{eff}}, \quad (25)$$

where $\bar{\omega}$ and κ_{eff} are defined below Eq. (17) and the cumulative noise

$$\begin{aligned} & \sqrt{\kappa_{\text{eff}}} \hat{b}_{\text{eff}}(t) \\ &= \sqrt{\kappa_m} \hat{b}_m(t) + ig^* e^{-i\Delta t} [\hat{W}_{r,\text{opt}}^\dagger(t) \hat{W}_{b,\text{opt}}(t) - \sqrt{N_b N_r} e^{i\mathcal{W}(t)}]. \end{aligned}$$

The statistics for $\kappa_{r,b} \gg \kappa_{\text{eff}}$: $\langle \hat{b}_{\text{eff}} \rangle = 0$, $\langle \hat{b}_{\text{eff}}^\dagger(t') \hat{b}_{\text{eff}}(t) \rangle \approx n_{\text{th}} \delta(t - t')$, and $\langle \hat{b}_{\text{eff}}(t) \hat{b}_{\text{eff}}^\dagger(t') \rangle \approx (n_{\text{th}} + 1) \delta(t - t')$,

$$n_{\text{th}} = \frac{\kappa_m n_{\text{eq}} + \bar{\kappa}_r}{\kappa_{\text{eff}}} \rightarrow \frac{n_{\text{eq}} + P_b/P_{\text{sat}}}{1 + (P_r - P_b)/P_{\text{sat}}}, \quad (26)$$

and \rightarrow holds for impedance-matched optical coupling $\kappa_{r,b} = K_{r,b}$. Equation (25) is equivalent to the equation of motion for magnons in equilibrium [Eq. (16) with $g = 0$] after substituting $\omega_m \rightarrow \omega_m + \bar{\omega}$, $\kappa_m \rightarrow \kappa_{\text{eff}}$, and $\hat{b}_m \rightarrow \hat{b}_{\text{eff}}$. Therefore in the steady state

$$\lim_{t \rightarrow \infty} \langle \delta \hat{M}^\dagger(t) \delta \hat{M}(t) \rangle = n_{\text{th}}, \quad (27)$$

justifying the notation n_{th} . At $P_b - P_r = P_{\text{sat}}$, the magnon damping κ_{eff} vanishes and the magnon number n_{th} diverges. The system becomes unstable and magnon nonlinearities should be taken into account [43]. For $T \sim 1$ K, $n_{\text{eq}} \sim 10$ and $n_{\text{th}} \sim n_{\text{eq}}$ for realistic powers $P_{r,b} \ll P_{\text{sat}}$. Thus, $n_{\text{th}} \ll n_c$, i.e.,

the coherently precessing magnetization is accompanied only by a small thermal cloud.

A large magnon population increases the BLS scattering cross section [21–23]: the uniform mode can be observed in BLS by exciting $> 10^{12}$ magnons by microwaves [22] in spite of the small optomagnonic coupling $g < 2\pi \times 5$ Hz. We consider now the enhancement of BLS by the high-momentum mode M that is coherently excited as discussed above. This can be measured by a third (probe) beam that couples to another optical WGM. Typically, only one of the sidepeaks dominates [16], with a ratio of scattered to incident (impedance-matched) photons

$$S = \frac{|g'|^2(n_c + n_{\text{th}})}{\kappa_{\text{opt}}^2}, \quad (28)$$

where g' is the coupling of the probe WGMs with the magnon mode M and κ_{opt} is a typical optical linewidth. For $g' = 2\pi \times 200$ Hz we require $P_{r,b} = 5$ mW for a signal that exceeds the noise background $S_{\text{noise}} \sim 10^{-5}$ [22]. A threefold larger $\{g, g'\}$ when reducing the radius to 100 μm increases S by two orders of magnitude (because $n_c \propto |g|^2$). For thin magnetic disks with $|g| = 2\pi \times 4$ kHz [17] $S \sim 1$.

Coherent magnons can also be excited by femtosecond laser pulses with a frequency spectrum that overlaps with the two WGMs, a process known as “impulsive stimulated Raman scattering” [14,26,44]. Time-periodic and phase-coherent laser pulses (frequency combs) [45,46] have a spectrum of sharp and periodic peaks whose period can be tuned to a magnon frequency. These techniques can achieve high laser intensities, but are less selective.

In summary, we show that two counterpropagating slightly detuned lasers can excite a large $\sim 10^6$ – 10^8 number of coherent magnons with submicrometer wavelengths in a conventional experimental setup of a proximity-coupled YIG sphere of radius ~ 300 μm . The consequent enhancement of the BLS cross section makes it experimentally feasible to observe. The coherent optical excitation of short-wavelength magnons with high group velocities can serve as an improved interface between light and spintronic devices in quantum domain.

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