Supporting Online Material for
Spin-Wave Lifetimes Throughout the Brillouin Zone

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Materials and Methods

The single-crystal MnF$_2$ sample, with volume of 8.5 cm$^3$, has extremely good mosaicity, which was measured by gamma-ray diffractometry to be $\leq 39^\circ$ FWHM. The crystal was mounted in the (H0L) scattering plane. The scattered neutron wave vector $k_f$ was held constant at 1.7, 2.15, or 2.5 Å$^{-1}$. The temperature was varied between 3 and 40 K using a closed-cycle cryostat. We measured the linewidths of magnons traveling in the $q_c$ direction, in the antiferromagnetic Brillouin zone centered at (100) ($S1$).

In a TAS experiment, the scattered neutrons which arrive at the detector lose energy and momentum which lie within an ellipsoid centered around a point in ($Q, \omega$)-space on the dispersion curve of the excitation. This “resolution ellipsoid” of the spectrometer has three dimensions in $Q$-space and one in energy. In a TAS experiment, the convolution of the resolution ellipsoid with the excitation surface in ($Q, \omega$)-space is measured. A scan in which the neutron energy transfer is varied (a “constant-Q scan”) reflects the spectral function in energy (e.g., a Lorentzian) which characterizes the dispersion curve. However, at low temperatures excitation linewidths are generally quite small, and the measured linewidth is thus dominated by the spectrometer resolution.

In a neutron resonance spin-echo TAS (NRSE-TAS) linewidth measurement, the resolution ellipsoid influences the measured linewidth only indirectly (by determining which neutrons reach the detector). The strategy of such a measurement is to manipulate the Larmor phase of each detected neutron such that this phase reflects only the value of the spectral function in energy of the corresponding excitation created by the neutron. This “focusing” technique ($S2$) can be applied exactly to excitations with linear dispersions. In this case, the detected neutrons which create excitations lying on a given line $\omega(q)$ parallel to the dispersion curve all have the same Larmor phase, independent of the momentum $q$ transferred by an individual neutron. Thus, a given value of the spectral function corresponds uniquely to a particular Larmor phase of the scattered neutrons. The Larmor phases of the individual neutrons together determine the polarization measured at the detector. As a consequence, the linewidth of the excitation can be

References
measured with high resolution.

The focusing is accomplished by rotating the RF resonance coil pairs independently around their vertical axes and by tuning the RF frequencies in the coil pairs. The appropriate orientation of the resonance coils for the measurement of a given magnon is

\[ \mathbf{n}_i \parallel (\bar{\mathbf{v}}_i - \nabla_q \omega_o), \]  

where \( \mathbf{n} \) is the unit vector perpendicular to the coil surface, \( i = 1,2 \) refers to the pair of coils in the spin-echo arm before and after the sample, respectively, \( \bar{\mathbf{v}} \) is the average neutron velocity in the respective spin-echo arm, and \( \nabla_q \omega_o \) is the slope of the dispersion curve of the magnon being measured. In this experiment, the coils were rotated by up to 30°; the limit of the instrument is 50°. The RF frequencies are tuned by setting the spin-echo time \( \tau \) (see Fig. 2) to be the same in both spin-echo arms. \( \tau \) is given by

\[ \tau = \frac{\omega_i L_i}{m (\bar{\mathbf{v}} \cdot \mathbf{n})_i [||\bar{\mathbf{v}}_i||^2 - \bar{\mathbf{v}}_i \cdot \nabla_q \omega_o]}, \]  

where \( \omega \) is the RF coil frequency and \( m \) is the neutron mass. \( L \) is the distance between pairs of resonance coils, which is approximately 0.5 m in each arm.

In the linewidth measurement, \( \tau \) is varied by changing the frequency in the RF coils, and the polarization of the detected neutrons is measured at each value (see Fig. 2). To perform a measurement of the polarization, one of the resonance coils is translated by a small amount (a total of a few millimeters), as a consequence of which the neutrons scattered from the sample describe a full cycle of their Larmor phases. The measured signal at the detector then reflects the depolarization of the scattered beam that results from the finiteness of the magnon linewidth. Fig. S1 shows such a “coil scan” taken with \( q = 0.2 \) r.l.u. at \( T = 15 \) K; the corresponding polarization of the scattered beam is 38.7%.

In order to shield the neutrons from external fields (including the Earth’s field) which would alter their Larmor phase, the path of the neutrons through the spin-echo arms and the sample is shielded by mu-metal boxes (see Fig. S1).

The raw linewidth data was then corrected for instrumental and non-intrinsic effects. Depolarization of the neutron beam resulting from instrumental effects was taken into account as a function of the RF coil frequency, the tilt angles of the spin-echo coils, and the neutron wavelength in each spin-echo arm. An analytical treatment of the non-intrinsic linewidth contribution which derives from second-order instrumental effects, the sample mosaicity, and the curvature of the dispersion, all of which contribute to dephasing of the Larmor phase over the
volume of the four-dimensional TAS resolution ellipsoid, was applied to the data (S3). Of these effects, the correction for the curvature of the magnon dispersion dominates, thanks to the excellent sample mosaicity; the former is largest for $q = 0$. For example, the $q = 0$ data taken at 25 K with $k_f=1.7$ Å$^{-1}$ corrects from $20.3 \pm 1.0 \mu eV$ to $11.7 \pm 1.0 \mu eV$. At larger $q$, the combined correction due to the above effects is not large. For example, for the data in Fig. 2, the raw data taken at 15 K and $q = 0.15$ is described by a linewidth of $15.6 \pm 0.9 \mu eV$, and the processed data by $14.9 \pm 0.9 \mu eV$, a change of 4%.
Fig. S1: Detected neutron intensity as a function of the position of the second RF resonance coil in the second spin-echo arm. This data was taken at $q = 0.2$ r.l.u. with $T = 15$ K, using an RF frequency of 127 kHz in the second spin-echo arm, which corresponds to $\tau = 38$ ps (see Fig. 2). The final neutron wave vector was fixed at $k_f = 1.7$ Å$^{-1}$. The counting time was roughly 200 s per point.
References

S1. We use a notation in which the momentum $Q$ transferred by the neutron is $Q = q + G$, where $q$ is the momentum transfer within the Brillouin zone centered at the reciprocal lattice vector $G$. These quantities are expressed in reciprocal lattice units (r.l.u.). For instance, $Q=\langle HKL \rangle$, with $Q_a = H(2\pi/a)$, and $q = \langle hkl \rangle$, with $q_c = l(2\pi/c)$.
