Magnetic excitations in weakly coupled spin dimers and chains material Cu$_2$Fe$_2$Ge$_4$O$_{13}$

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Magnetic excitations in a weakly coupled spin dimers and chains compound Cu$_2$Fe$_2$Ge$_4$O$_{13}$ are measured by inelastic neutron scattering. Both structure factors and dispersion of low energy excitations up to 10 meV energy transfer are well described by a semiclassical spin wave theory involving interacting Fe$^{3+}$ ($S = 5/2$) chains. Additional dispersionless excitations are observed at higher energies, at $\omega_0 = 24$ meV, and associated with singlet-triplet transitions within Cu$^{2+}$-dimers. Both types of excitations can be understood by treating weak interactions between the Cu$^{2+}$ and Fe$^{3+}$ subsystems at the level of the Mean Field/Random Phase Approximation. However, this simple model fails to account for the measured temperature dependence of the 24 meV mode.

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I. INTRODUCTION

The ground states of low-dimensional magnets are strongly affected by quantum spin fluctuation. In so-called quantum spin liquids spin correlation remains short-range even at zero temperature, and the excitation spectrum is gapped. This disorder is relatively robust and such systems resist long-range ordering even in the presence of residual 3D interactions or anisotropy. In contrast, gapless low-dimensional magnets are very sensitive to external perturbations that can easily drive them towards long-range ordering. New physics is found in bicomponent systems that combine these two distinct types of low-dimensional spin networks. An example is $R_2$BaNiO$_5$ materials where Haldane spin chains weakly interact with magnetic rare earth ions.\textsuperscript{1,2,3} More recently, we reported the discovery and study of a novel quantum ferrimagnet Cu$_2$Fe$_2$Ge$_4$O$_{13}$\textsuperscript{4,5} We showed that this compound can be viewed as a system of antiferromagnetic (AF) Cu-dimers that weakly interact with almost classical Fe$^{3+}$ chains. This weak coupling leads to a rather unusual cooperative ordering phase transition at low temperatures.

The crystal structure of Cu$_2$Fe$_2$Ge$_4$O$_{13}$ is monoclinic $P2_1/m$ with $a = 12.101$ Å, $b = 8.497$ Å, $c = 4.869$ Å, and $\beta = 96.131^\circ$.\textsuperscript{6} The arrangement of magnetic ions and likely exchange pathways is shown in Fig. 1(a). Fe$^{3+}$ ions form crankshaft-shaped chains that run in the $b$ direction. These chains are separated by Ge$_4$ tetrahedra in $c$ direction. The Cu$^{2+}$ dimers are located in-between Fe$^{3+}$ chains along the $a$ direction. Simultaneous cooperative long-range magnetic ordering of the two magnetic subsystems occurs at 40 K.\textsuperscript{5} The magnetic structure is roughly collinear, with spins lying in the crystallographic $a - c$ plane. In addition, there are small out-of-plane spin components (canting). The saturation magnetic moment on Cu$^{2+}$ ions is anomalously small, being suppressed by residual quantum fluctuations in the dimers: $m_{\text{Cu}} = 0.38(4)\mu_B$. This suggests that the coupling $J_{\text{Cu-Fe}}$ between the Cu-dimers to the Fe-subsystem is weak compared to intra-dimer AF interactions $J_{\text{Cu}}$. The pairs of Cu-spins remain in a spin-singlet state that is only partially polarized by interactions with the long-range order in the Fe-subsystem. The data collected in preliminary inelastic neutron scattering experiments for energy transfers up to 10 meV could be well explained by fluctuations of Fe$^{3+}$ spins alone. The Fe$^{3+}$ spins form weakly-coupled $S = 5/2$ chains, the corresponding effective exchange constants being $J_{\text{Fe}} = 1.60(2)$ meV, $J'_{\text{Fe}} = 0.12(1)$ meV, as shown in Fig. 1. These values are consistent with rough estimates of exchange constants based on magnetic susceptibility data: $J_{\text{Fe}} = 1.7$ meV, $J_{\text{Cu}} = 25$ meV. To date, no excitations associated with the Cu-dimers could be identified.

While the very nature of the crankshaft-shaped chains implies some alternation of bond strength, we find no evidence thereof in the measured dispersion curves. In fact, they are well reproduced by a model involving uniform magnetic Fe-chains with weak interchain interactions along the $c$ direction. In the remainder of this work we shall therefore disregard the bond-alternation and assume the Fe-chains to be magnetically uniform.

In the present paper we report a more detailed inelastic neutron scattering study of Cu$_2$Fe$_2$Ge$_4$O$_{13}$. Our main re-
sult is an observation of a complete separation of energy scales for Cu$^{2+}$ and Fe$^{3+}$-centered magnetic excitations. An analysis of the intensity pattern for low-energy spin waves allows us to unambiguously associate them with the dynamics of $S = 5/2$ chains. In addition, separate narrow-band excitation originating from Cu subsystem is observed at higher energy transfers. The layout of the paper will be as follows. In section III we will describe the characteristics of our samples and experimental setups. Section III describes the results for low-energy excitations in single-crystal samples, as well as measurements of the much weaker high energy excitations in a large-volume powder sample. In Section IV we shall interpret the observed separation of energy scales by a Mean Field-Random Phase Approximation (MF-RPA) treatment of interactions between Cu$^{2+}$- and Fe$^{3+}$ spins. Discussion and conclusion will be drawn in Section V and VI, respectively.

II. EXPERIMENTAL

High quality single crystals with the dimension of $3 \times 4 \times 35$ mm$^3$ were grown by the floating zone method. All samples were found to be twinned. The two twins share a common $(b, c)$ plane, the monoclinic structure allowing two independent orientations of the $a$ axis. In reciprocal space the domains share a common $(a^*, b^*)$ plane, but have distinct $c^*$ axes, as illustrated in Fig. II (b). Two separate single crystals were co-aligned to obtain a larger sample of cumulative mosaic spread $0.44^\circ$. For single crystal inelastic neutron scattering experiments we exploited four different setups. Setup I employed the SPINS cold neutron spectrometer at the NIST Center for Neutron Research (NCNR). The scattering plane was defined by the $b^*$ and the bisector $c'*$ of the $c^*$ axes in the two crystallographic domains, as shown in Fig. II (b). In this geometry the scattering planes are the same in both domains. It is convenient to define $c^* = c' \cos(\beta - 90^\circ)$. The momentum transfer in the scattering plane of the spectrometer is then indexed by Miller indexes $h'$, $k'$ and $l'$ of a fictitious orthorhombic structure. For the two domains $h = \pm (c^*/a^*)l' \tan(\beta - 90^\circ), h = k'$ and $l = l'$. Since $\beta$ is close to $90^\circ$, $h$ is almost zero for most measurements using Setup 1, where $l$ is small. In Setup 2 the scattering plane was $(a^*, b^*)$, which is also common for the two domain types. In both setups we used (guide) $-80' - 80' - (open)$ collimation with a Be filter positioned after the sample and a fixed final energy $E_f = 5$ meV or 3 meV. The data were collected at $T = 1.4$ K using a standard He-flow cryostat. Setup 3 was used for wide surveys in reciprocal space and employed the HB1 thermal neutron spectrometer at the High Flux Isotope Reactor at ORNL. The scattering plane was $(a^*, b^*)$, as in Setup 2. The collimation was $48' - 40' - 40' - 240'$. Neutrons with $E_f = 13.5$ meV were used in conjunction with a Pyrolytic graphite (PG) filter positioned after the sample. The experiments in Setup 3 were performed at $T = 6.4$ K maintained by a closed-cycle He refrigerator.

Intensity was the main limiting factor for studies of higher-energy magnetic excitations. To maximize sample volume the measurements were performed on a 50 g polycrystalline Cu$_2$Fe$_2$Ge$_4$O$_{13}$ powder that was prepared by the solid state reaction method. This experiment was performed on the HB3 3-axis spectrometer at HFIR with $48' - 40' - 60' - 120'$ collimations and a PG filter after the sample (Setup 4). The final neutron energy was fixed at $E_f = 14.7$ meV. A closed-cycle refrigerator was used to achieve low temperatures.

III. EXPERIMENTAL RESULT

A. Low energies

In this section we concentrate on the low energy excitations at energy transfers up to 10 meV. As was explained
FIG. 2: Energy scans collected in Cu$_2$Fe$_2$Ge$_4$O$_{13}$ for momentum transfers $\mathbf{q} = (0 \ k \ 0.5)$. White and grey circles correspond to measurements with $E_f = 5$ and 3 meV, respectively. Solid lines are Gaussian fits.

in our preliminary report and will be discussed in detail below, this part of the spectrum can be associated with conventional spin waves from the Fe-subsystem. The Cu-dimers only provide an effective Fe-Fe interaction, but contribute nothing to the dynamics at low energies.

Typical energy scans at $\mathbf{q} = (0 \ k \ 0.5)$ measured using setup 1 are shown in Fig. 2. White and grey circles correspond to data collected with $E_f = 5$ meV and 3 meV respectively. Well defined resolution-limited peaks are observed in the entire Brillouin zone. Solid lines are Gaussian fits to the data after a subtraction of a linear background. The excitation energy is a minimum at the magnetic zone center at $\mathbf{q} = (0 \ 2k \ 0.5)$. A small gap of about 1.6 meV is observed at this wave vector. The apparent shoulder structure can be attributed to a splitting of the spin wave branch into two components with somewhat different gap energies. The gaps are most likely anisotropy-related. The zone-boundary energy at $\mathbf{q} = (0 \ 3.0 \ 0.5)$ is about 9 meV. The dispersion relation was obtained by Gaussian fits to individual scans and is shown in Fig. 3 (a). The $k$-dependence of the measured energy-integrated peak intensity is plotted in symbols in Fig. 3 (b). The intensity scales roughly as $1/\omega$.

Energy scans at $\mathbf{q} = (0 \ 2.0 \ l)$ and $(0 \ 2.9 \ l)$ are shown in Fig. 4. Constraints on experimental geometry prevented us from reaching the more symmetric $(0 \ 3.0 \ l)$ reciprocal-space rods at higher energy transfers. These two sets of data reveal two distinct excitation branches, that are strongest near even and odd $k$-values, respectively. The former branch is dispersive along the $c$ axis, while the latter one is almost flat. Dispersion relations and the integrated intensity plots for both modes were obtained using Gaussian fits and are shown in Fig. 5 (a) and (b). Even for the more dispersive lower-energy branch the boundary energy along the $c^*$ direction is only about 5 meV: about half of that in the $b^*$ direction.

No dispersion of low-energy excitations could be detected along the $a^*$ direction. This is illustrated by the energy scans collected on the $(h \ 2.0 \ 0)$ reciprocal-space...
rod using Setup 2 (Fig. 6). Two peaks are observed at 5 and 9 meV, respectively. The excitation energies are plotted in Fig. 7 (a). Interestingly, the peak intensity is strongly dependent on $h$, as shown in Fig. 7 (b).

From the data presented above one can immediately conclude that the most relevant magnetic interactions are within the Fe$^{3+}$ layers, parallel to the $(b,c)$ plane. Inter-layer coupling along the $a$-direction is considerably weaker. Based on structural considerations, it must involve the Cu$^{2+}$ spins.

**B. Higher-energy excitations**

The model that we previously proposed for Fe$_2$Cu$_2$Ge$_4$O$_{13}$ (Ref. 5) implies a separation of energy scales of spin wave like excitations on the Fe subsystem and triplet excitations of the Cu dimers. From magnetic susceptibility measurements we have estimated the intra-dimer exchange constant to be around 25 meV. Due to small sample size, in single crystal experiments we failed to observe any clear magnetic inelastic features in this energy transfer range. To search for the Cu-triplet mode we performed additional measurements on a large-size powder sample.

The powder data collected at $T = 12$ K are summarized in the false color plot in Fig. 8 (a). The data were obtained by combining 17 separate constant-$q$ scans. For each such scan a linear background was subtracted from the measured intensity. Figure 8 (a) clearly shows a narrow excitation band at $\hbar \omega \sim 24$ meV. The measured intensity of the 24 meV peak decreases with the increase of $q$, as expected for magnetic scattering. The observed energy width at $q = 2.3$ Å$^{-1}$ is somewhat larger than experimental resolution, but still small compared to the central energy (Fig. 8 (b)). Such energy dependence in powder samples typically indicates a narrow dispersion bandwidth (see, for example, Refs. 6,7). The observed spectrum is consistent with the excitations originating from the structural Cu-dimers, indicated by the thick solid bonds in Fig. 1 (a). However, the momentum transfer range covered in the experiment, especially at $|q| \rightarrow 0$ is insufficient for a more detailed analysis of the structure factor. In particular, the size of the dimers can not be
independently extracted from the experimental data.

To obtain additional information on the 24 meV excitation we studied its temperature dependence at \( q = 2.3 \, \text{Å}^{-1} \). The main challenge was dealing with a large background that originates from (i) temperature-independent scattering, including spurious scattering from an “accidental” Bragg powder line at 24.8 meV and (ii) temperature-dependent phonon scattering. These two contributions can be removed from the data if one assumes that the useful magnetic signal is relatively weak and/or temperature-independent above \( T = 200 \, \text{K} \). This would indeed be true if the magnetic scattering originated from effectively isolated Cu-dimers with an intradimer exchange constant of 24 meV. The phonon contribution, which is assumed to scale with the Bose factor, is thus estimated from comparing scans at \( T = 300 \, \text{K} \) and \( T = 200 \, \text{K} \). It is then appropriately scaled and subtracted from all scans, leaving only the true magnetic signal and the \( T \)-independent background. The two can not, in principle, be reliably separated. However, we can follow the change in magnetic signal from \( T = 200 \, \text{K} \) by using the phonon-subtracted \( T = 200 \, \text{K} \) scan as “background”.

The result of this elaborate background subtraction is plotted in Fig. 8. The well-defined peak seen at low temperature broadens progressively with increasing \( T \) and practically disappears at \( T \gtrsim 40 \, \text{K} \). In this range there also seems to be a downward shift in the peak’s central energy.

IV. ANALYSIS

A. Separation of energy scales

To understand the dynamics of the coupled \( \text{Fe}^{3+} \) and \( \text{Cu}^{2+} \) subsystems we shall make the central assumption that the exchange constant \( J_{\text{Cu}} \) that binds pairs of \( \text{Cu}^{2+} \) spins into AF dimers is the largest energy scale in the sys-

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FIG. 6: Energy scans measured in \( \text{Cu}_2\text{Fe}_2\text{Ge}_4\text{O}_{13} \) for momentum transfers \( q = (h \, 2.0 \, 0) \). Symbols and lines are as in Fig. 4.

FIG. 7: (a) Measured energies (a) and integrated intensities (b) of magnetic excitations as a function of momentum transfer along the \( \alpha^* \) direction. Symbols and lines are as in Fig. 5.
FIG. 8: (a) False-color plot of inelastic intensity measure $d$ in Cu$_2$Fe$_2$Ge$_4$O$_{13}$ powder samples as a function of energy and momentum transfer. (b) Typical energy scans measured at $T = 12$ K for different momentum transfers. Heavy solid lines are Gaussian fits. The shaded Gaussians represent experimental energy resolution.

Under these circumstances the degrees of freedom associated with Cu-spins can be effectively integrated out at low energies. Indeed, the isolated Cu$^{2+}$-subsystem has a spin singlet ground state and a large energy gap $\Delta = J_{\text{Cu}}$. At $\hbar \omega \ll \Delta$ it lacks any intrinsic dynamics, i.e., its dynamic susceptibility is purely real and almost energy-independent. In the spirit of RPA, from the point of view of Fe$^{3+}$ spins, the Cu-dimers merely act as a polarizable medium that can transfer magnetic interactions between the Fe-layers. The staggered spin susceptibility of each $S = 1/2$ dimer being $2/(J_{\text{Cu}})$, this effective coupling, labeled as $J_{\text{eff}}$ in Fig. 1, is given by:

$$J_{\text{eff}} = \frac{J_{\text{Cu}}^2}{J_{\text{Cu}} - \text{Fe}}.$$  \hspace{1cm} (1)

Thus, to a good approximation, the low-energy spin dynamics of Cu$_2$Fe$_2$Ge$_4$O$_{13}$ is simply that of the Fe$^{3+}$-subsystem with an additional exchange coupling. From the experiment, where no dispersion of spin waves could be observed along the $a$-axis, the effective exchange constant must be rather small. Nevertheless, as explained in our previous paper it is absolutely crucial in completing a 3-dimensional spin network and allowing long-range magnetic ordering at a non-zero temperature.

At high energy transfers, comparable to the Cu-dimer gap, it is the Fe$^{3+}$ degrees of freedom that can be effectively integrated out. At $T < T_N$ their effect is reduced to producing a static staggered exchange spin field that acts on the Cu$^{2+}$ spins and is proportional to the ordered Fe$^{3+}$ moment:

$$h_{\text{Cu}} = \langle S_{\text{Fe}} \rangle J_{\text{Cu-Fe}}.$$  \hspace{1cm} (2)

In our approximation at high energy transfers Cu$_2$Fe$_2$Ge$_4$O$_{13}$ behaves as a collection of (possibly interacting) Cu-dimers in an effective staggered field $h_{\text{Cu}}$.

B. Spin waves and dynamic structure factor for low energies

As explained above, the low-energy spectrum of Cu$_2$Fe$_2$Ge$_4$O$_{13}$ can be understood by considering the Fe-subsystem in isolation, and even $J_{\text{eff}}$ can be ignored for its smallness. For the magnetically ordered state the dynamic structure factor of such a system can be calculated.
\[ S(q, \omega) = S_0(q, \omega) \cos^2 2\pi \delta_a h \cos^2 2\pi \delta_b k + \frac{1}{2} S_0(q + (0, 1, 0), \omega)(1 + \sin^2 2\pi \delta_a h \sin^2 2\pi \delta_b k) \]
\[ + S_0(q + (0, 2, 0), \omega) \cos^2 2\pi \delta_a h \sin^2 2\pi \delta_b k + \frac{1}{2} S_0(q + (0, 3, 0), \omega)(1 + \sin^2 2\pi \delta_a h \sin^2 2\pi \delta_b k), \quad (3a) \]

\[ \delta_a = 0.1239, \delta_b = 0.0629. \quad (3b) \]

The SWT dynamic structure factor for a collinear antiferromagnet on a Bravais lattice is well known:\(^7\)

\[ S_0(q, \omega) = (u_q + v_q)^2 \delta(h\omega_b - \omega) \quad (4a) \]
\[ u_q^2 = \frac{S(h\omega_b + 2Sj(0))}{h\omega_b}, \quad (4b) \]
\[ u_qv_q = \frac{-2S^2j(q)}{h\omega_b}, \quad (4c) \]
\[ h\omega_q = S\sqrt{j(0)^2 - j(q)^2 + \Delta^2}. \quad (4d) \]

Here \(j(q)\) is the Fourier transform of exchange interactions, and \(\Delta\) empirically accounts for the anisotropy gap. In our particular case of the Fe\(^{3+}\) subsystem in Cu\(_2\)Fe\(_2\)Ge\(_4\)O\(_{13}\) we have:

\[ j(q) = 2(J_{Fe} \cos \frac{\pi}{2} k + J'_{Fe} \cos 2\pi l), \quad (5) \]

The cross section for inelastic neutron scattering from spin waves is given by:

\[ \frac{d^2 \sigma}{d\Omega dE} \propto |F(q)|^2 \left[ 1 + \left( \frac{q_y}{q} \right)^2 \right] \langle n_q + 1 \rangle S(q, \omega), \quad (6) \]

In this formula \(F(q)\) is the magnetic form factor for Fe\(^{3+}\), \(\langle n_q \rangle\) is the Bose factor and \(q_z\) is the projection of the scattering vector onto the direction of ordered Fe\(^{3+}\) moments.

C. Fits to data

The model cross section given by Eq. (6) can accurately reproduce the observed low-energy spectra in Cu\(_2\)Fe\(_2\)Ge\(_4\)O\(_{13}\). Due to the presence of four terms in Eq. (5), there are four distinct spin wave branches, that we shall denote as modes I through IV, correspondingly. The dispersion relation given by Eq. (4d) was fit to the experimental data shown in Figs. 3 (a) and 5 (a) using a least-squares algorithm. A good fit is obtained with \(J_{Fe} = 1.60\) meV, \(J'_{Fe} = 0.12\) meV, and \(\Delta = 2.02\) meV. The result is shown in lines in figures 3 (a) and 5 (a). With these parameters our model also agrees well with the measured dispersion (or, rather, absence thereof) along the \(a\) axis, as shown in 4 (a).

What is important, is that not only the energies, but also the intensities of the observed excitations are well reproduced by our model. Calculated intensities for each mode or combined intensities of a couple of modes in cases where experimental energy resolution is insufficient to resolve individual branches, are shown in lines in Figs. 3 (a), 5 (a) and 7 (a). This quantitative agreement between the measured and observed structure factors confirms that the scattering is indeed due to Fe\(^{3+}\) spins. An excellent illustration of this was obtained by mapping out...
FIG. 11: (a) Constant energy scans at $\hbar\omega = 8.5$ meV in wide $q = (h \, k \, 0)$ range. (b) Simulation based of our model cross section convoluted with the experimental resolution function.

the scattering intensity in a wide $q$-range using Setup 4, as shown in Fig. 11 (a). These data correspond to a fixed energy transfer $\hbar\omega = 8.5$ meV, and have a characteristic checkerboard pattern. Our spin wave model with the parameters quoted above reproduces this behavior very well, as shown in Fig. 11 (b). In this calculation the model cross section was numerically convoluted with the known experimental resolution function. The apparent periodicity along $k$ is due to a steep dispersion that takes the excitations in and out of the probed energy range. However, the periodicity along $h$ is related to the trigonometric coefficients in Eq. (3a). These, in turn, are determined by the geometry of the crankshaft-shaped Fe$^{3+}$ chains in Cu$_2$Fe$_2$Ge$_4$O$_{13}$.

V. DISCUSSION

As demonstrated above, the low-energy spin dynamics of Cu$_2$Fe$_2$Ge$_4$O$_{13}$ is well described by an effective spin wave theory for the Fe$^{3+}$ spin chains. Based on the available data it is impossible to unambiguously associate the observed 24 meV mode with the Cu$^{2+}$ dimers. However, much confidence in this assumption can be drawn from a recent study of Cu$_2$Sc$_2$Ge$_4$O$_{13}$. In this isostructural compound, only the Cu$^{2+}$ ions are magnetic. Indeed the magnetic susceptibility showed $S = 1/2$-dimers behavior with similar energy to Cu$_2$Fe$_2$Ge$_4$O$_{13}$.

We can now estimate all the relevant exchange interactions in the system. Using Eq. (2) in combination with the staggered susceptibility of an isolated antiferromagnetic dimer, from the known saturation moments $\langle S_{\text{Cu}} \rangle = 0.18(8)$ and $\langle S_{\text{Fe}} \rangle = 1.77(4)$ for Cu$^{2+}$ and Fe$^{3+}$, respectively, we get $J_{\text{Cu-Fe}} = 2.54(3)$ meV. The effective coupling is then $J_{\text{eff}} = 0.13(4)$ meV. The results for all exchange parameters are summarized in Table I.

Our model for Cu$_2$Fe$_2$Ge$_4$O$_{13}$ is qualitatively consistent with the observed slight increase of the energy of the Cu-dimer mode with decreasing temperature. Indeed, the gap energy of isolated dimers, as that of other gapped systems, is known to increase with the application of a staggered field. In Cu$_2$Fe$_2$Ge$_4$O$_{13}$ this field is generated by the ordered moment on the Fe$^{3+}$ sites, and at $T < T_N$ increases proportionately to $\langle S_{\text{Fe}} \rangle$. Beyond that, the observed $T$-dependence of the 24 meV mode is different from that for isolated dimers. In the latter, the intensity would remain almost constant below $T = 40$ K. Moreover, the peak would remain sharp at all temperatures. The discrepancy may be related to intrinsic limitations of the MF/RPA approach, and merits further investigation. In particular, it is tempting to somehow associate the observed emergence and sharpening of the 24 meV inelastic peak with the onset of long-range order.

VI. CONCLUSION

Our data bring solid quantitative support to the concept of separation of energy scales in the mixed-spin quantum antiferromagnet Cu$_2$Fe$_2$Ge$_4$O$_{13}$. Using a simple MF/RPA approach we are able to determine all the relevant exchange interactions. However, certain features of the temperature dependence of spin excitations require further theoretical and experimental study.

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TABLE I: Exchange parameters for Cu$_2$Fe$_2$Ge$_4$O$_{13}$.

| $J_{Fe}$  | $J'_{Fe}$ | $J'_{eff}$ | $J_{Cu-Fe}$ | $J_{Cu}$  |
|----------|-----------|------------|-------------|-----------|
| 1.60(2) meV | 0.12(1) meV | 0.13(4) meV | 2.54(3) meV | 24.(2) meV |

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