Localized magnetic excitations in the fully frustrated dimerized magnet Ba$_2$CoSi$_2$O$_6$Cl$_2$

Nobuyuki Kurita$^1$, Daisuke Yamamoto$^2$, Takuya Kanaseka$^2$, Nobuo Furukawa$^2$, Seiko Ohira-Kawamura$^3$, Kenji Nakajima$^3$, and Hidekazu Tanaka$^1$

$^1$Department of Physics, Tokyo Institute of Technology, Oh-okayama, Meguro-ku, Tokyo 152-8551, Japan
$^2$Department of Physics and Mathematics, Aoyama-Gakuin University, Sagamihara, Kanagawa 252-5258, Japan
$^3$Materials and Life Science Division, J-PARC Center, Tokai, Ibaraki 319-1195, Japan

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A coupled spin dimer system, in which antiferromagnetic (AF) dimers interact with one another through interdimer exchange interactions, provides an opportunity for correlating condensed matter physics with particle physics. One intriguing feature is the crystallization of magnetic quasiparticles, magnons (or triplons), like a Wigner crystal, which could be a key to understanding exotic quantum phases such as supersolids and flat-band solid states. The crystallized phase of magnons is expected to emerge under a magnetic field when the frustration of the interdimer exchange interactions is so strong that magnons become localized. This quantum phenomenon can be characterized by a stepwise magnetization process and dispersionless magnetic excitations. Until recently, experimental studies have been, to our best knowledge, limited to the Shastry–Sutherland compound SrCu$_2$(BO$_3$)$_2$ and Ba$_2$CoSi$_2$O$_6$Cl$_2$. In this compound, magnons are localized owing to the orthogonal configuration of dimers. Fractional magnetization plateaus observed in SrCu$_2$(BO$_3$)$_2$ imply the successive crystallization of magnons.

The ground state of a spin dimer system is typically a spin singlet with an excitation gap $\Delta$ to the lowest excited triplet state. When a magnetic field exceeding the critical field $H_c(=\Delta/g\mu_B)$ is applied, magnons are created on the dimer lattice. Magnons can hop to neighboring dimer sites and interact with each other via transverse and longitudinal components of interdimer exchange interactions, respectively. For the simplified two-dimensional (2D) case, as illustrated in Fig. 1(a), the hopping and repulsive terms are proportional to $(J_{11} + J_{22}) - (J_{12} + J_{21})$ and $J_{11} + J_{22} + J_{12} + J_{21}$, respectively. Given that the frustration of interdimer exchange interactions is perfect, namely $J_{11} + J_{22} = J_{12} + J_{21}$, the hopping of magnons becomes completely suppressed and magnons form a periodic array consisting of half-filled magnons owing to the competition between the repulsive interactions of magnons and the Zeeman energy. When the hopping term is dominant, by contrast, the spin dimer system undergoes an XY-type AF ordering upon applying a magnetic field of above $H_c$. It is known that a magnetic-field-induced quantum phase transition can be described by the Bose–Einstein condensation (BEC) of magnons.

Magnetic excitations of the effective spin $S = 1/2$ dimerized magnet Ba$_2$CoSi$_2$O$_6$Cl$_2$ have been probed directly via inelastic neutron scattering experiments at temperatures down to 4K. We observed five types of excitation at 4.8, 5.8, 6.6, 11.4, and 14.0 meV, which are all dispersionless. The scattering intensities of the three low-lying excitations were found to exhibit different $Q$-dependences. Detailed analysis has demonstrated that Ba$_2$CoSi$_2$O$_6$Cl$_2$ is a two-dimensional spin dimer system where the interdimer exchange interactions are almost perfectly frustrated and the undimerized spins, even slightly included in crystals, make an essential contribution to the excitation spectrum.

Recently, the magnetic insulators Ba$_2$CoSi$_2$O$_6$Cl$_2$ and Ba$_2$CuSi$_2$O$_6$Cl$_2$ were reported to be a new series of 2D spin dimer systems with the exchange network shown in Fig. 1(a). High field magnetization measurements of Ba$_2$CoSi$_2$O$_6$Cl$_2$ up to 70T revealed the complete magnetization process with a magnetization plateau at half of saturation magnetization $M_s$, and the edges of the magnetization plateau reported for SrCu$_2$(BO$_3$)$_2$ are rather smeared, the 1/2$M_s$ plateau observed in Ba$_2$CoSi$_2$O$_6$Cl$_2$ is sharply stepwise. This suggests that interdimer exchange interactions are almost perfectly frustrated. For a definitive conclusion, it is important to elucidate the magnetic excitations of this compound. Ba$_2$CoSi$_2$O$_6$Cl$_2$ crystallizes in a monoclinic structure with the space group $P2_1/c$. The lattice parameters are $a = 7.1382$ Å, $b = 7.1217$ Å, $c = 18.6752$ Å, and $\beta = 91.417^\circ$. Owing to the strong spin orbit coupling and pyramid-like crystal field, the effective spin of magnetic Co$^{2+}$ ions can be described by an $S = 1/2$ strongly XY-like XXZ model at temperatures much lower than the spin-orbit coupling constant of $|\lambda|/k_B \sim 250$ K.

In this letter, we report on inelastic neutron scattering measurements of Ba$_2$CoSi$_2$O$_6$Cl$_2$. The observed dispersionless excitations, together with the results of previous magnetization measurements, provide strong evidence for the almost perfect frustration of the interdimer exchange interactions. It was found that undimerized spins cre-
Incident neutron energies at J-PARC, Japan [25]. The measurements with two sets of chopper spectrometer AMATERAS installed in the Materials and Life Science Experimental Facility (MLF) at J-PARC, Japan [25]. The measurements with two sets of incident neutron energies $E_i = (2.6, 5.9, 10.5, 23.6)\text{ meV}$ and $(2.9, 4.7, 7.7, 15.2)\text{ meV}$ were performed at several temperatures of 4K to 240K. Approximately 60 plate-like single crystals with a total mass of $\sim 1\text{ g}$ were glued on an aluminum plate, where the $a$ axis (or $b$ axis) for each crystal was aligned parallel to the horizontal direction. Note that the single crystals used in this study were twinned, where the $a$ and $b$ axes were interchanged. The fluoropolymer (CYTOP®) employed as the glue had a negligible contribution to the background. The wave vector $k_i$ of an incident neutron was set parallel to the $c^*$ axis. All the data were analyzed using the software suite Utsusemi [27].

Figure 2(a) shows the energy-momentum map of the scattering intensity along $Q = (Q_a, 0, 0)$ at the base temperature of 4K measured with an incident neutron energy of $E_i = 15.2\text{ meV}$, where the scattering intensity is integrated over $Q_b$ and $Q_c$. One can confirm three strong dispersionless excitations at 4.8, 5.8, and 6.6 meV and two weak dispersionless excitations at higher energies of 11.4 and 14.0 meV, as shown in Figs. 2(e)–(g). With increasing temperature above 150 K, the excitation spectrum shown in Fig. 2(a) is considerably smeared and the intensity decreases. From this result, the origin was verified to be magnetic. Figures 2(e)–(g) show scattering intensities as a function of energy measured at 4K with $E_i = 7.7, 15.2,$ and 23.6 meV, respectively, where scattering intensities were integrated over whole $Q_a, Q_b$, and $Q_c$ ranges. Since the width of the excitations at 4.8, 5.8, 6.6, and 11.4 meV are resolution-limited, all the excitation peaks except that at 14.0 meV are single peaks and not the superposition of two or more excitation peaks.

As shown in Fig. 2(h), the scattering intensity oscillates along $Q_c$. This oscillation is related to the spin separation $R$ in the dimer. The dynamical structure factor $S(Q, \omega)$ is proportional to $\sin^2 ((Q \cdot R)/2)$. Because in Ba$_2$CoSi$_2$O$_6$Cl$_2$, the spin separation $R$ is approximately parallel to the $c$ direction, the oscillation of intensity occurs along $Q_c$ but does not along $Q_a$ and $Q_b$. The wave vector $Q^\text{max}$ that gives the local maxima of the intensity which is proportional to $S(Q, \omega)f^2(Q)$, where $f(Q)$ is the magnetic form factor of Co$^{2+}$, is calculated to be $Q^\text{max} = (3.0, 9.0 \cdots)\text{ meV}$. These values are consistent with the experimental results shown in Fig. 2(h). Note that the decrease in scattering intensities for $1.8 < Q_a < 2.5$ in Fig. 2(a) is caused by the neutron absorption mainly owing to the plate-shaped samples and the sample holder of the aluminum plate, and thus is extrinsic.

The noteworthy feature of the three low-energy excitations at 4–7 meV is that the scattering intensities exhibit different $Q$-dependences. This is more evident in the constant-energy slices of the scattering intensity shown in Figs. 2(b)–(d), where the scattering intensity is integrated over $Q_c$, considering good two-dimensionality as evidenced by the observed dispersionless excitations along $Q_c$ [Fig. 2(h)]. The middle excitation with the highest intensity at 5.8 meV is nearly independent of $(Q_a, Q_b)$. On the other hand, the intensities of the upper-side (6.6 meV) and lower-side (4.8 meV) excitations exhibit local maxima when both $Q_a$ and $Q_b$ are integers and half-integers, respectively. For the excitations at 11.4 and 14.0 meV, the low intensities make it difficult to discern their $Q$-dependence and to explain why the peak width of the excitation at 14.0 meV is much broader than the calculated resolution limit. The energies of the single singlet-triplet excitation to the $|t_{\pm 1}\rangle$ and $|t_0\rangle$ states are given by $E_1 = (J^1 + J^2)/2$ and $E_2 = J^1$, respectively. In Ba$_2$CoSi$_2$O$_6$Cl$_2$, the energy level of $|t_0\rangle$ is higher than that of $|t_{\pm 1}\rangle$ owing to the strong XY anisotropy [8]. It is considered that when an excited triplet is localized, the single singlet-triplet excitation is dispersionless and its...
intensity is independent of \( Q_a \) and \( Q_b \). Thus, the middle excitation peak at 5.8 meV can be assigned to the single singlet-triplet excitation to the \( |t_{\pm 1}\rangle \) state. Assuming that the excitation at 11.4 meV corresponds to the single singlet-triplet excitation to the \( |t_0\rangle \) state, we obtain \( J_{\perp} = 11.4 \) meV and \( J_{\parallel} = 0.16 \) meV. This means that the intradimer exchange interaction closely approximates the XY model.

Because there is no other single-triplet excitation to the \( |t_{\pm 1}\rangle \) states, we can deduce that all the dimers are magnetically equivalent. Thus, the sharply stepwise magnetization process with a 1/2-magnetization plateau can only be described in terms of the crystallization of localized magnons owing to the strong frustration of interdimer exchange interactions \( S \). From these observations, we can safely conclude that the intradimer interactions in \( \text{Ba}_2\text{CoSi}_2\text{O}_6\text{Cl}_2 \) almost satisfy the perfect frustration condition \( J_{11}^{\perp} + J_{22}^{\perp} = J_{12}^{\perp} + J_{21}^{\perp} \). Note that the sharp side peaks observed at 4.8 and 6.6 meV do not indicate the presence of the multiple dimer sites with different magnitudes of the intradimer exchange interaction \( J_{\perp} \), because their intensities exhibit different periodicities that are commensurate with \( a^* \) and \( b^* \). As shown below, these side peaks rather support the perfect frustration scenario with a single dimer site.

Here we discuss the origin of the anomalous side peaks observed at 4.8 and 6.6 meV. For simplification, we assume that \( J_{11}^{\perp} = J_{22}^{\perp} = J_{12}^{\perp} = J_{21}^{\perp} = J_{1}^{\perp} \) and \( J_{12}^{\parallel} = J_{21}^{\parallel} = J_{1}^{\parallel} \). Given that the \( \text{Ba}_2\text{CoSi}_2\text{O}_6\text{Cl}_2 \) crystals employed in this study are perfect crystals, the side-peak structure will be absent from the excitation spectrum. It is natural to assume that these side peaks are produced by interdimer interactions, because the energy difference between these side peaks and the middle peak is on the order of the interdimer interactions and the intensities of these side peaks are commensurate with \( a^* \) and \( b^* \). One plausible scenario is that the observed side peaks are caused by the three-body problem among dimer spins and a neighboring undimerized single spin produced by a vacancy of \( \text{Co}^{2+} \), as illustrated in Fig. 1(c).

The model Hamiltonian of the three-body problem is written as

\[
\mathcal{H}_{3b} = J_1^{\perp}(S_1^x S_2^x + S_1^y S_2^y) + J_2^{\parallel} S_1^z S_2^z \\
+ \tilde{J}_1^{\perp}(S_1^x S_3^x + S_1^y S_3^y) + \tilde{J}_1^{\parallel} S_1^z S_3^z \\
+ \tilde{J}_2^{\perp}(S_2^x S_3^x + S_2^y S_3^y) + \tilde{J}_2^{\parallel} S_2^z S_3^z,
\]

where \( J_1^{\perp} \) and \( \tilde{J}_2^{\perp} \) are the exchange interactions between \( S_3 \) and \( S_1 \) and between \( S_3 \) and \( S_2 \), respectively, which are in general different from \( J_2^{\parallel} \) and \( \tilde{J}_1^{\parallel} \) of the host system and no longer identical to each other, \( \tilde{J}_1^{\perp} \neq \tilde{J}_2^{\perp} \). The ground (\( n = 0 \)) and \( n \)th excited states \( |\psi_n\rangle \) are expressed as

\[
|\psi_n(S^z = \frac{1}{2})\rangle = u_n|s, \uparrow\rangle + v_n|t_1, \downarrow\rangle + w_n|t_0, \uparrow\rangle,
\]

\[
|\psi_n(S^z = -\frac{1}{2})\rangle = u_n|s, \downarrow\rangle - v_n|t_{-1}, \uparrow\rangle - w_n|t_0, \downarrow\rangle,
\]

for (2)
for \( n = 0, 1 \) and \( 3 \), while for \( n = 2 \)

\[
|\psi_2(S^z = -\frac{3}{2})rangle = |t_{-1}, \downarrow\rangle.
\]

(3)

Here, \( \uparrow \) and \( \downarrow \) represent the spin states of the undimerized spin \( S_3 \). The coefficients \( \{u_n, v_n, w_n\} \) \( (n = 0, 1, \) and \( 3) \) can be chosen to be real without loss of generality. The eigenstates of each energy level are doubly degenerate and labeled by the \( z \) component of the total spin \( S^z \). The excitation energies of the side peaks observed at 4.8 and 6.6 meV and the weak excitation observed at 14.0 meV are reproduced by the set of parameters \( (\tilde{J}_{11}^\perp, \tilde{J}_{12}^\perp, \tilde{J}_{12}^\parallel, \tilde{J}_{22}^\perp) = (7.93, 0.66, 3.08, 0.25) \) meV, which gives \( u_0 = 0.936, v_0 = 0.333, w_0 = -0.116, u_1 = -0.349, v_1 = 0.827, w_1 = -0.442, u_3 = -0.051, v_3 = 0.454, \) and \( w_3 = 0.890 \). Here, we used the naive assumption that the exchange anisotropy in \( \tilde{J}_{12}^{\perp} \) is identical to that in \( \tilde{J}_{22}^{\perp} \), i.e., \( \tilde{J}_{11}^{\perp} / \tilde{J}_{12}^{\perp} = \tilde{J}_{12}^{\parallel} / \tilde{J}_{22}^{\perp} \). The discussion below is qualitatively independent of this simplification. Note that the other set of parameters obtained by interchanging \( \tilde{J}_{11}^{\perp} \) and \( \tilde{J}_{22}^{\perp} \), i.e., \( (\tilde{J}_{11}^\parallel, \tilde{J}_{12}^\perp, \tilde{J}_{12}^\parallel, \tilde{J}_{22}^\perp) = (3.08, 0.25, 7.93, 0.66) \) meV, which gives \( u_0 = 0.936, v_0 = -0.333, w_0 = 0.116, u_1 = 0.349, v_1 = 0.827, w_1 = -0.442, u_3 = 0.051, v_3 = 0.454, \) and \( w_3 = 0.890 \), also gives the same excitation energies. However, only the case of \( \tilde{J}_{11}^{\perp} > \tilde{J}_{22}^{\perp} \) can reproduce the \( Q \) dependence of the scattering intensities as shown below.

In the above-mentioned scenario, one undimerized spin affects the local excitations of the eight neighboring spins as can be seen in Fig. 1. From the low-temperature Curie tail observed in the susceptibility data, the amount of undimerized spins in the Ba$_2$CoSi$_2$O$_6$Cl$_2$ single crystals is estimated to be 3~4\% of all spins. Hence, it is considered that the undimerized spins make a contribution of \( \sim 30\% \) to the excitation spectrum. This could be the reason for the high intensity of the side peaks observed at 4.8 and 6.6 meV.

To explain the periodic intensity oscillations in the excitations at 4.8 and 6.6 meV, we calculate the differential cross section of the inelastic neutron scattering via the effective three-body Hamiltonian given by Eq. (1) using the exchange parameters obtained above. The character of the scattering intensity for the transition from the initial state \( |\psi_0\rangle \) to an excited state \( |\psi_n\rangle \) is mainly determined by the exclusive structure factor tensor, which is expressed as

\[
S_{\alpha\beta}^{(0\rightarrow n)}(q) = \sum_{ij} e^{iq(\mathbf{r}_i - \mathbf{r}_j)} \langle \psi_0 | \hat{S}_i^\alpha | \psi_n \rangle \langle \psi_n | \hat{S}_j^\beta | \psi_0 \rangle,
\]

(4)

with \( \alpha, \beta = x, y, z \). Since the \( z \) component of the total spin, \( S^z \), is a conserved quantity, the exclusive structure factor tensor is given in the diagonal form \( S_{\alpha\beta}^{(0\rightarrow n)}(q) = S_{\alpha\alpha}^{(0\rightarrow n)}(q) \delta_{\alpha\beta} \). Here we choose the relative coordinates among the three spins \( \hat{S}_1, \hat{S}_2, \) and \( \hat{S}_3 \) as \( \hat{r}_{12} = (0, 0, c) \), \( \hat{r}_{31} = (\pm a, 0, 0) \), and \( \hat{r}_{32} = (\pm a, 0, c) \), respectively. Note that the result does not depend on the total spin \( S^z = \pm \frac{1}{2} \) of the initial state \( |\psi_0\rangle \).

For the scattering accompanied by the energy transfer \( E_n - E_0 = 4.8 \) and 6.6 meV for \( n = 1 \) and \( 2 \), respectively, the exclusive structure factors are written as

\[
S_{\alpha\alpha}^{(0\rightarrow n)}(q) = A_n^\alpha + B_n^\alpha \cos q_x c + C_n^\alpha \cos q_z a + D_n^\alpha \cos(q_x a + q_z c),
\]

\( (5) \)

\[
S_{zz}^{(0\rightarrow n)}(q) = A_n^\parallel + B_n^\parallel \cos q_x c + C_n^\parallel \cos q_z a + D_n^\parallel \cos(q_x a + q_z c),
\]

\( (6) \)

where the coefficients are given \( u_n, v_n, \) and \( w_n \). Since the scattering intensity shown in Figs. 2(b)–(d) is integrated over \( Q_c \), the main contribution to the oscillation of the intensity along the \( Q_a \) direction is from the term \( \frac{1}{2} C_n^\alpha \cos q_z a \). Note that the oscillation along the \( Q_b \) direction can be explained in the same manner from the effective three-body problem of a dimer and its neighboring undimerized spin in the \( b^* \) direction. The coefficients \( C_n^\alpha \) are given by

\[
C_1^\alpha = -\sqrt{2}(u_0 v_1 + v_0 u_1 + v_1 w_1 + w_0 v_1)(u_0 v_1 - w_0 w_1),
\]

\[
C_1^\parallel = 2(u_0 v_1 + w_0 v_1 + v_0 u_1)(u_0 v_1 - v_0 u_1 + w_0 w_1),
\]

\[
C_2^\parallel = -\sqrt{2}(u_0 - w_0 v_1), \quad C_3^\parallel = 0,
\]

\[
C_4^\parallel = -\sqrt{2}(u_0 v_3 + v_0 u_3 + v_1 w_3 + w_0 v_3)(u_0 u_3 - w_0 w_3),
\]

\[
C_3^\parallel = 2(u_0 v_3 + w_0 v_3 + v_0 u_3)(u_0 u_3 - v_0 u_3 + w_0 w_3).
\]

(7)

A crucial factor to produce the \( Q \)-dependent oscillation of the scattering intensities of the side peaks is a mixture of the singlet and triplet states in the \( \psi_{01} \) states owing to the entanglement with the undimerized spin state. The oscillating terms vanish without the mixing of singlet and triplets in the \( \psi_{01} \) states. Using the values of \( u_n, v_n, \) and \( w_n \) derived from the set of parameters \( (\tilde{J}_{11}^\perp, \tilde{J}_{12}^\perp, \tilde{J}_{12}^\parallel, \tilde{J}_{22}^\perp) = (7.93, 0.66, 3.08, 0.25) \) meV, we obtain \( C_1^\perp = 0.221, C_1^\parallel = 0.108, C_2^\perp = -0.495, \) and \( C_2^\parallel = 0 \). These coefficients correctly reproduce the \( Q \)-dependent oscillation of the scattering intensities, which exhibit local maxima when both \( Q_a \) and \( Q_b \) are integers and half-integers for the excitations at 4.8 and 6.6 meV, respectively. The weak excitation observed at 14.0 meV is expected to be assigned to the transition from the \( \psi_0 \) state to the \( \psi_3 \) state. Since we obtain \( C_1^\perp = -0.051 \) and \( C_3^\parallel = -0.598 \), the scattering intensity should also show a \( Q \)-dependent oscillation with local maxima when
$Q_a$ and $Q_b$ are both half-integers. However, because of the small intensity, it is difficult to distinguish the $Q_a$ (or $Q_b$) dependence of its intensity in the experimental data. Note that using the parameter set obtained by exchanging $J_1^{\perp\perp}$ and $J_2^{\perp\perp}$, the values of $C_{a}^{\perp\parallel}$ become $C_{a}^{\perp\parallel} = -0.480$, $C_{b}^{\parallel\perp} = -0.713$, $C_{2}^{\perp\parallel} = 0.386$, and $C_{3}^{\perp\parallel} = 0$, which are inconsistent with the experimental data.

To conclude, we have probed the magnetic excitations of Ba$_2$CoSi$_2$O$_6$Cl$_2$ directly via inelastic neutron scattering measurements. The five observed types of magnetic excitation are dispersionless and exhibit characteristic $Q$-dependences of scattering intensities. It was found that the excitation spectra can reasonably be explained not only by a "perfect frustration" scenario for interdimer interactions in the host system, but also by emergent three-body quantum states owing to undimerized spins included by vacancies in the crystals. To obtain further experimental findings to increase understanding of this system, it is important to elucidate the field evolution of each excitation by, for example, in-field neutron scattering and electron spin resonance experiments.

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