**Electronic Supplementary Information**

**Novel copper-azido magnetic molecular tapes: syntheses, structures, and magnetic properties**

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**Structure refinement special details of complex 3:**

Five restraints were used in the structure refinement of complex 3. Details are provided below:

\texttt{dfix 1.40 0.01 C1 N20 C3 N21}
\texttt{dfix 1.10 0.01 N17 N18 N17 N18'}
\texttt{delu 0.002 N1 N2}

Refinement without these restraints yielded an unstable and unsuitable refinement. Due to the sharp shift of the bond lengths of C1–N20 and C3–N21, the distances between C1 and N20 atoms, C3 and N21 atoms, are restrained to a target value 1.40 with an estimated standard deviation 0.01. The distances between N17 and N18 atoms, N17 and N18’ atoms are restrained to a target value 1.10 with an estimated standard deviation 0.01, due to the minor disorder of the N16–N17–N18 moiety. The DELU restraint is applied to N1–N2 distance, due to the larger effective standard deviation.

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Fig. S1 PXRD patterns of 1 as-synthesized sample and simulated one based on the single-crystal structures.

Fig. S2 PXRD patterns of 2 as-synthesized sample and simulated one based on the single-crystal structures.
**Fig. S3** PXRD patterns of 3 as-synthesized sample and simulated one based on the single-crystal structures.

**Fig. S4** Temperature dependence of in-phase ($\chi_M'$) and out-of-phase ($\chi_M''$) ac magnetic susceptibility for 1 obtained zero external magnetic field, $H_{ac} = 5$ Oe, 10 Hz. The solid line is a guide for the eye.
**Fig. S5** Temperature dependence of in-phase (χ′) and out-of-phase (χ″) ac magnetic susceptibility for 2 obtained zero external magnetic field, $H_{ac} = 5$ Oe, 10 Hz. The solid line is a guide for the eye.

**Fig. S6** Temperature dependence of in-phase (χ′) and out-of-phase (χ″) ac magnetic susceptibility for 3 obtained zero external magnetic field, $H_{ac} = 5$ Oe, 10 Hz. The solid line is a guide for the eye.
Fig. S7 Magnetization versus field up to $H = 70 \text{ kOe}$ at 1.8 K for 1.

Fig. S8 Magnetization versus field up to $H = 70 \text{ kOe}$ at 1.8 K for 2.
Fig. S9 Magnetization versus field up to $H = 70$ kOe at 1.8 K for 3.