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### Magnetic phase diagram of F<sub>2</sub>PNNNO<sup>1</sup>

YASU TAKANO, University of Florida

2-[2',6',-difluoro-4'-(*N-tert*-butyl-*N*-oxyamino)phenyl]-4,4,5,5-tetramethyl-4,5-dihydro-1*H*-imidazol-1-oxyl 3-oxide, F<sub>2</sub>PNNNO for short, is an organic molecule containing two unpaired electrons. Residing in the *N-tert*-butyl nitroxide and nitronyl nitroxide groups, the two  $S=1/2$  spins of these electrons are ferromagnetically coupled with an exchange constant of 407 K. In a crystal, two neighboring F<sub>2</sub>PNNNO molecules form a pair in which the nitronyl-nitroxide spins are coupled antiferromagnetically with an exchange constant of 67 K. The magnetism of F<sub>2</sub>PNNNO is that of the spin tetramers of these molecular pairs, which in turn are antiferromagnetically coupled with an exchange constant of 7.4 K [1]. Specific-heat and magnetocaloric-effect measurements reveal a highly symmetric boundary of the ordered phase in the phase diagram, with a lower critical field of  $H_{c1}=9.46$  T and an upper critical field of  $H_{c2}=15.37$  T. The ordering temperature  $T_c$  obeys a power law  $T_c \sim (H - H_{c1})^\alpha$  near  $H_{c1}$ , with the exponent  $\alpha$  approaching 2/3 in the low-temperature limit, indicative of a Bose-Einstein condensation (BEC) of  $|S, S_z \rangle = |1,1\rangle$  tetramers. Near the upper critical field  $H_{c2}$ , where one expects a BEC of singlet  $|0,0\rangle$  tetramers in the “vacuum” comprising  $|1,1\rangle$  tetramers, the corresponding power-law exponent remains around 0.4. Remarkably, the temperature dependence of the specific heat indicates that the magnon dispersion is independent of magnetic field between the two critical fields. This work is in collaboration with H. Tsujii, B. Andraka, Y. Hosokoshi, and K. Inoue. [1] Y. Hosokoshi *et al.*, *Phys. Rev. B* **60**, 12924 (1999).

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