Reply to “Comment on ‘Transition from Bose glass to a condensate of triplons in Tl$_{1-x}$K$_x$CuCl$_3’”’

Fumiko Yamada$^1$, Hidekazu Tanaka$^1$, Toshio Ono$^1$, and Hiroyuki Nojiri$^2$

$^1$Department of Physics, Tokyo Institute of Technology, Meguro-ku, Tokyo 152-8551, Japan
$^2$Institute for Material Research, Tohoku University, Aoba-ku, Sendai 980-8577, Japan

(Dated: June 16, 2011)

Showing low-temperature specific heat and other experimental data and also on the basis of established physics, we argue against the comment made by Zheludev and H"uvonen criticizing our recent study on the magnetic-field-induced spin ordering and critical behavior in Tl$_{1-x}$K$_x$CuCl$_3$, which is described as the Bose glass-condensate transition of triplons.

PACS numbers: 72.15.Rn, 75.10.Jm, 75.40.Cx, 76.30.-v

In our recent paper$^1$ referred to as paper I, we reported specific heat and ESR studies of the magnetic-field-induced Bose glass to Bose-Einstein condensate transition of triplons and the critical behavior in Tl$_{1-x}$K$_x$CuCl$_3$. However, Zheludev and H"uvonen criticize that the specific heat peak shown in Fig. 1(b) in paper I is rounded; thus, the ambiguity of the transition field $H_N$ is $\pm 0.2$ T at low temperatures and leads to the different critical exponents $\phi = 1.74$ and 1.39 for $x = 0$ and 0.36, respectively. Here, the critical exponent $\phi$ is defined as

$$H_N(T) - H_c = AT^\phi,$$

where $H_N(T)$ and $H_c$ are the transition fields at finite and zero temperatures, respectively. They also criticize that the specific heat peak in Tl$_{1-x}$K$_x$CuCl$_3$ does not diverge at $H_N$; thus, the transition is not a continuous thermodynamic transition but a crossover. They ascribe the absence of the true phase transition to the staggered $g$-tensor and the Dzyaloshinsky-Moriya (DM) antisymmetric interaction. In what follows, we argue against their criticisms.

Actually, the low-temperature specific heat data shown in Fig. 1(b) in paper I appear shrunk, because we plotted many field scan data measured at various temperatures. In Fig. 1, we show the increase in the specific heat vs magnetic field measured at 0.45 K. Although the specific heat peak is rounded, we can determine the transition field $H_N$ within an error of $\pm 0.05$ T, which is approximately the same as the size of symbols in Figs. 1(c) and 2 in paper I. The error in determining $H(T)_N$ is not as large as $\pm 0.2$ T.

For $x \neq 0$, the transition field $H(T)_N$ increases rapidly with temperature. The effect of the error on the estimation of the critical exponent is small. As shown in Fig. 2 in paper I, the low-temperature phase boundary for $x \neq 0$ cannot be described by a single exponent $\phi$, although Comment authors claim that the phase boundary can be expressed by the single exponent $\phi = 1.39$. To investigate the change in the exponent with a fitting window, we fit eq. (1) in the temperature range of $T_{\text{min}} \leq T \leq T_{\text{max}}$, setting the lowest temperature at $T_{\text{min}} = 0.36$ K and varying the highest temperature $T_{\text{max}}$ from 1.87 to 0.82 K. We actually observed a systematic decrease in the exponent $\phi$ with decreasing $T_{\text{max}}$, as shown in Fig. 2 in paper I.

Comment authors claim that the phase boundaries for both $x = 0$ and $x \neq 0$ can also be expressed by the same critical exponent $\phi = 1$. It is obvious from Fig. 1(c) in paper I that the phase boundary for pure TlCuCl$_3$ is perpendicular to the field axis, while those for $x \neq 0$ are not. Because of large three-dimensional interdimer interactions of the order of 1 meV, magnetic excitations in TlCuCl$_3$ are largely dispersive in three dimensions. This leads to a small triplon mass and a small coefficient $A$ in eq. (1). For this reason and a large saturation field of $H_s \simeq 90$ T, the temperature-field region of $T < 2$ K and $H - H_c < 1$ T can be considered as a critical region for pure TlCuCl$_3$, which is close to the quantum critical point (QCP). Near the QCP, the transition field $H(T)_N$ does not exhibit a large temperature dependence.
FIG. 2: Magnetic field dependence of magnetic peak intensity in Tl$_{1-x}$K$_x$CuCl$_3$ with $x = 0.17$ for (0, 0, 1) measured at 0.5 K.

Because for pure TlCuCl$_3$, the transition field $H_N(T)$ below 1 K scarcely depends on temperature and the error in determining $H_N(T)$ from the field scan of specific heat is $\pm 0.05$ T, it is insufficient to obtain a correct exponent only from data points below 1 K. For this reason, we used data points up to 2 K and obtained $\phi = 1.53$. This analysis should be appropriate.

Actually, the specific heat peak shown in Fig. 1 is rounded. This should be ascribed to the instrumental resolution. Comment authors strongly suspect that the field-induced transition in Tl$_{1-x}$K$_x$CuCl$_3$ is smeared by an antisymmetric interaction, such as the Zeeman term with the staggered $g$ tensor and the DM interaction, and thus, the transition is not a true phase transition. Indeed, it is difficult to assume that there is no antisymmetric interaction in Tl$_{1-x}$K$_x$CuCl$_3$, but we can estimate the upper limit of the magnitude of the antisymmetric interaction. The magnitude of the anisotropy can be evaluated from the ESR linewidth $\Delta H$, which is given by the magnetic anisotropy that does not commute to the total spin. Near 0.5 K, the ESR line shape is between Lorentzian and Gaussian, which indicates that exchange narrowing is less effective. In such a case, the ESR linewidth corresponds to the magnitude of the local field due to magnetic anisotropy. From $\Delta H \simeq 0.1$ T near 0.5 K, the total energy $\Delta E$ of the anisotropy including the staggered Zeeman term and the DM interaction can be evaluated as $\Delta E \simeq 0.01$ meV. The magnitude $\Delta E_{AS}$ of the antisymmetric interaction is smaller than $\Delta E$, and thus, $\Delta E_{AS}$ is much smaller than the intradimer interactions of the order of 1 meV. Therefore, we consider that the effect of the antisymmetric interaction on the field-induced transition is negligible.

Neutron diffraction data may be useful for determining whether the transition is well-defined. At present, we have diffraction data only for $x = 0.17$. In Fig. 2, we show the field dependence of the (0, 0, 1) magnetic reflection for $x = 0.17$ measured at 0.5 K in magnetic fields parallel to the $b$ axis. A clear bend anomaly due to the field-induced phase transition is observed at $H_N = 4.2$ T. The sharpness of the bend anomaly for $x = 0.17$ is similar to that in the case of pure TlCuCl$_3$.

Comment authors claim that specific heat diverges at a continuous transition point. However, this seems to be a misunderstanding on their part. It is established that the critical exponent $\alpha$ for specific heat is negative for the three-dimensional XY and Heisenberg universality classes, and thus, the specific heat does not diverge at the transition points for these cases.

1. F. Yamada, H. Tanaka, T. Ono, and H. Nojiri, Phys. Rev. B 83, 020409(R) (2011).
2. A. Zheludev and D. H¨ uvonnen, Phys. Rev. B. in press.
3. N. Cavadini, G. Heigold, W. Henggeler, A. Furrer, H.-U. Güdel, K. Kr¨ amer, and H. Mutka, Phys. Rev. B 63, 172414 (2001).
4. A. Oosawa, T. Kato, H. Tanaka, K. Kakurai, M. Müller, and H.-J. Mikeska, Phys. Rev. B 65, 094426 (2002).
5. M. Matsumoto, B. Normand, T. M. Rice, and M. Sigrist: Phys. Rev. B. 69, 054423 (2004).
6. T. Nikuni, M. Oshikawa, A. Oosawa, and H. Tanaka, Phys. Rev. Lett. 84, 5868 (2000).
7. G. Misguich and M. Oshikawa, J. Phys. Soc. Jpn. 73, 3429 (2004).
8. J. H. Van Vleck, Phys. Rev. 74, 61 (1948).
9. H. Tanaka, A. Oosawa, T. Kato, H. Uekusa, Y. Ohashi, K. Kakurai, and A. Hoser: J. Phys. Soc. Jpn. 70, 939 (2001).
10. J. C. Le Guillou and J. Zinn-Justin, Phys. Rev. Lett. 39, 95 (1977).