$YbCo_2$: Large Magnetic Entropy Change per Volume in Yb-based Metallic Magnetic Refrigerants for Sub-Kelvin Temperature

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A Yb-based intermetallic compound $YbCo_2$ exhibiting a magnetic-field-induced order is known to show a giant specific heat divided by temperature, $C/T \sim 6.5$ J/K^{2}mol, around 0.3 K. We investigate the potential of this substance as the magnetic refrigerants by measuring the thermodynamic properties down to 0.1 K. The entropy change per volume by applying magnetic field of 3 T is found to be $-\Delta S_{\rm M} \sim 0.15$ J/K cm^3 around 1 K. This value is particularly large in the Yb-based metallic magnetic refrigerants to provide the sub-Kelvin temperature. In addition, we demonstrate that a 2.2 gram sample is cooled down to 0.26 K by the adiabatic demagnetization refrigeration from 1.8 K and 12 T. This cooling performance is attributed to the ground state, at zero field, locating around the boundary between the field-induced ordered phase and the paramagnetic one. Our study clarifies the utility of the sub-Kelvin magnetic refrigerants which combine the large $-\Delta S_{\rm M}$ and the high thermal conductivity of metal.

Adiabatic demagnetization refrigeration (ADR) is a technique to cool down by demagnetizing the magnetic refrigerants on the quasi-adiabatic environments. Since this technique needs only a magnet and magnetic materials as the refrigerants, we can reach the cryogenic temperature below 1 K without complex refrigerators using precious ^{3}He. Though paramagnetic salts including 3d-transition metals have been used as the cryogenic magnetic refrigerants for decades, it is not convenient to use them because of the chemical instability[1,2]. In recent years, various stable magnetic refrigerants including rare-earth ions have been studied. The low magnetic-transition temperature $T_{\rm M}$ in rare-earth ions retains the magnetic entropy used for magnetic refrigeration around absolute zero.

The performance as the magnetic refrigerants is evaluated by two parameters of the *adiabatic temperature change*, $\Delta T_{\rm ad}$, and the *magnetic entropy change*, $-\Delta S_{\rm M}$. The former is defined as $\Delta T_{\rm ad} = T_{\rm i} - T_{\rm f}$, where $T_{\rm i}$ ($T_{\rm f}$) indicates the initial (final) temperature before (after) demagnetization in the ADR experiments. The value of $-\Delta S_{\rm M}$ is evaluated as the difference between the entropy under the zero field and the magnetic field. We often need to mount the magnetic refrigerants on the narrow space in a refrigerator. In addition, the spatial area where the magnetic field is applied is limited. Therefore, the magnitude of $-\Delta S_{\rm M}$ per volume is of importance for the real application.

Main route to increase the amount of $-\Delta S_{\rm M}$ is focusing on the Gd-based compounds since a Gd^{3+} has an entropy of $\ln 8 = 3\ln 2$ arising from the spin $S = 7/2$. This value is 3 times as large as that in the ground state doublet realized in most other rare-earth ions affected by the crystalline electric field. Indeed, some magnetic refrigerants of Gd-based oxides/fluoride where $T_{\rm M}$ is suppressed by the geometrical frustration effect exhibit large $-\Delta S_{\rm M}$ exceeding 0.1-0.2 J/K cm^3 [3–9]. Instead of such a large $-\Delta S_{\rm M}$, if we demand $T_{\rm f}$ below ~ 0.1 K, the Yb-based oxides should be selected[9–12].

Another significant point to cool down to below 1 K is the thermal conductivity of the magnetic refrigerants because the thermal conductivity in the insulators mentioned above sharply decays as a power of temperature on cooling. In this point, Yb-based intermetallic compounds are advantageous as the cryogenic magnetic refrigerants to cool down to ~ 0.2 K[13–16]. The strongly localized nature of $4f$ electrons in the heavy rare-earth ion Yb^{3+} ($4f^{13}$) results in the low $T_{\rm M}$ related to $T_{\rm f}$. Another advantage peculiar to the strongly correlated metals including Yb or Ce is that $T_{\rm f}$ can be further lowered with the chemical substitution by tuning the ground state to the quantum critical points where $T_{\rm M}$ is absolute zero[17–19].

Thus, Yb-based intermetallic compounds are advantageous as the cryogenic magnetic refrigerants because of the low $T_{\rm f}$ and the high thermal conductivity. In this paper, we report the performance of $YbCo_2$ as magnetic refrigerants. $YbCo_2$ is one of the C15 Laves-phase compounds with the cubic $MgCu_2$-type structure[20]. The specific heat divided by temperature, C/T, divergently increases on cooling and exhibits a maximum at ~ 0.3 K, where the value of C/T is ~ 6.5 J/K^{2}mol [21,22]. A magnetically ordered state appears to set in at 0.3 K as inferred from the zero-field μSR experiments. This giant C/T and the density of Yb, accounting for 1/3 in the constituent atoms, are expected to provide the sizable entropy per volume at zero field . Notably, $YbCo_2$ exhibits a field-induced order above 1 T and 2 K[21]. In this field-induced phase, both moments in Yb and Co order magnetically[22]. Since the field-induced order takes away the entropy under magnetic field around absolute zero, enhancement of $-\Delta S_{\rm M}$ is expected.

Polycrystalline samples of $YbCo_2$ were prepared from Yb ingots and Co powder. An open tungsten crucible containing

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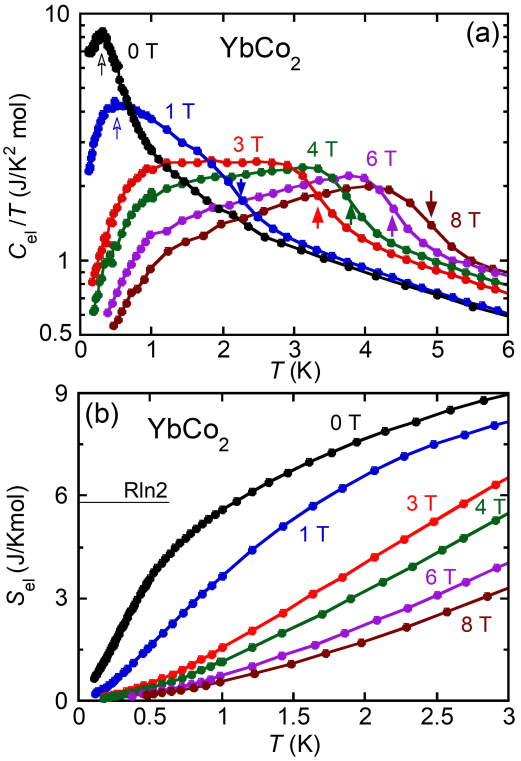

FIG. 1. (a) Electronic specific heat divided by temperature C_{el}/T under the magnetic field up to 8 T in $YbCo_2$. The closed and open arrows show the drops in C_{el}/T on heating by the field-induced phase transition and the peak, respectively. (b) Electronic entropy S_e derived from C_{el}/T.

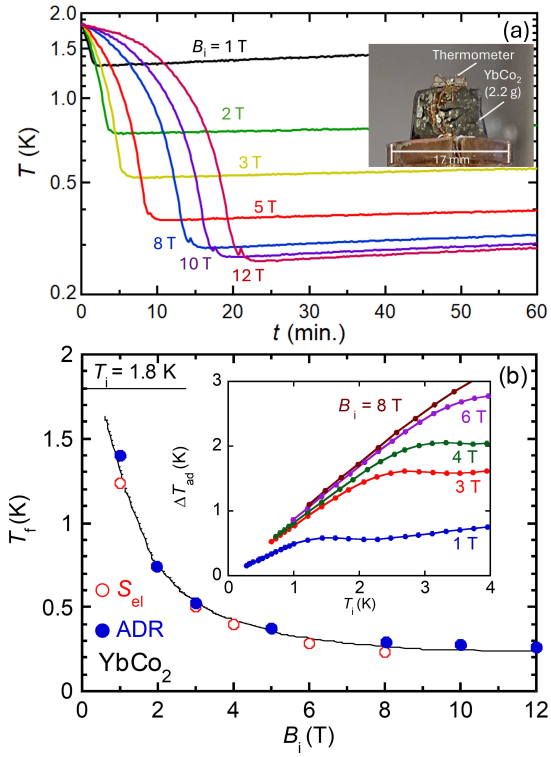

FIG. 2. (a) Time dependence of temperature in $YbCo_2$ by ADR from the initial temperature of $T_i = 1.8$ K and the various initial fields of $B_i = 1$, 2, 3, 5, 8, 10, and 12 T. The inset shows the picture of the sample used in this ADR experiment. This sample was mount on the laboratory-made ADR cell attachable to PPMS[19]. (b) Final temperature T_f from $T_i = 1.8$ K as a function of B_i. T_f are determined by the electronic entropy S_{el} in Fig. 1(b) (Open Circles) and the ADR experiment (Closed Circles). The inset displays the adiabatic temperature change $\Delta T_{ad} = T_i - T_f$ which is derived from S_{el} in Fig. 1(b).

them was encapsulated in a quartz ampoule under Ar atmosphere. This crucible in this quartz ampoule was heated up by a induction furnace. Eventually, $YbCo_2$ were obtained by reacting the Co powder with the melted Yb for 20-30 minutes. The cubic $MgCu_2$-type structure was confirmed by the X-ray powder diffraction. The ratio of Yb : Co in the sample were 1 : 2.03(1) determined by the electron-probe microanalysis.

Specific heat was measured down to 0.4 K by a ^{3}He option of PPMS (Quantum Design Co.) by the thermal relaxation method. Measurements at lower temperatures down to 0.1 K were conducted by using a laboratory-made calorimeter installed in the commercial Cambridge Magnetic Refrigerator mFridge in the magnetic field up to 4 T. Magnetization was measured down to 1.8 K and up to 5 T by MPMS (Quantum Design Co.). We measured magnetization down to 0.4 K by a capacitive Faraday magnetometer installed in a ^{3}He single-shot refrigerator (Heliox, Oxford Instruments)[23,24]. The ADR experiments were performed by a laboratory-made ADR cell attachable to PPMS. The setup and the procedure of our ADR experiments are described in Ref. [19].

Figure 1(a) shows the electronic specific heat divided by temperature, C_{el}/T, as a function of temperature in the magnetic field up to 8 T. C_{el} is obtained as $C_{el} = C - C_{nuc} - C_{ph}$, where C, C_{nuc}, and C_{ph} are the raw data of the specific heat, nuclear, and phonon contributions, respectively. In Fig. S1 of the Supplementary Material, we plot C/T and C_{ph}/T where C_{ph} extract from Ref. [21] is the specific heat of the non-magnetic reference compound YNi_2. In this paper, we focus on the low-temperature range below 6 K where C_{ph} is ignorable small, compared with C_{el}. In the magnetic fields up to 4 T, we observed a convex downward curvature in C/T below ~ 0.3 K as shown in Fig. S1. This curvature following $C_{nuc}/T \sim 1/T^3$ is subtracted from C/T.

As shown by the solid arrows in Fig. 1(a), a drop in C_{el}/T on heating is observed above 1 T and 2 K. As observed and discussed in Ref. [21, 22]. this drop is due to the field-induced phase transition whose phase boundary is plotted in the inset of Fig. 3(b). In addition, as indicated with the open arrows in Fig. 1(a), a sharp peak suggesting another phase transition is observed at 0.3 K at zero field. This peak is proposed to be the magnetic order because the internal field is observed below 0.4 K in the muon spin resonance experiments[22]. Figure 1(b) displays the temperature dependence of the electronic entropy S_{el} derived by integrating $C_{el}(T)/T$ with respect to the

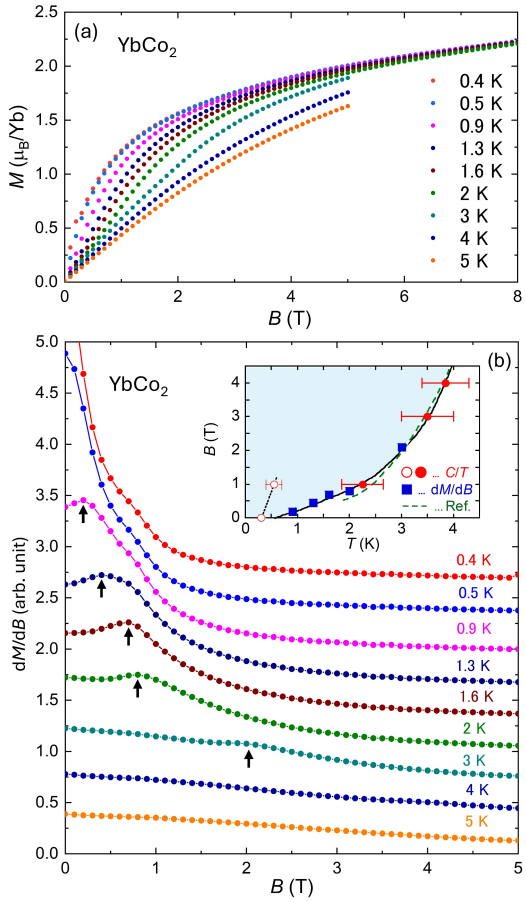

FIG. 3. (a) Magnetic field dependence of magnetization M down to 0.4 K up to 8 T in $YbCo_2$. (b) Field derivative of magnetization dM/dB. The peaks by the metamagnetic transition are indicated by the arrows. The inset shows the magnetic phase diagram constructed from the peaks in dM/dB (squares), the peaks (open circles) and the drops (closed circles) observed in $C_{\rm el}/T$ in Fig. 1(a). The dashed green line shows the phase boundary reported in Ref. [21].

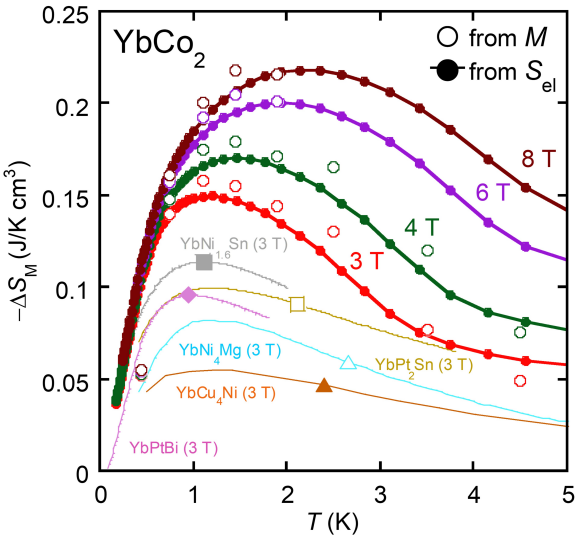

FIG. 4. The magnetic entropy change per volume $-\Delta S_{\rm M}$ as a function of temperature, determined by the magnetization M in Fig. 3 and the electronic entropy $S_{\rm el}$ in Fig. 1. For comparison, we also plot $-\Delta S_{\rm M}$ in the reported Yb-based metallic magnetic refrigerants for sub-Kelvin temperature under the magnetic field of 3 T[13–16,25]. In $YbPt_2Sn$, the average of entropy at 2 T and 4 T is adopted as entropy at 3 T[13].

temperature. At zero field, $S_{\rm el}(T)$ reaches $R\ln 2$ J/Kmol at $\sim$ 1 K resultant from the Kramers doublet of a Yb^{3+} ion. As described later, $-\Delta S_{\rm M}$ and $\Delta T_{\rm ad}$ are obtained from $S_{\rm el}$ at zero field and the magnetic field.

Figure 2(a) displays the time dependence of the sample temperature in the ADR experiments from $T_{\rm i} = 1.8$ K with various initial fields up to $B_{\rm i} = 12$ T. Demagnetization from t = 0 was performed with a speed of 0.6 T/min. The $YbCo_2$ sample with 2.2 gram mass was mount on a Cu-stage which is thermally insulated from the heat bath at 1.8 K as shown in the inset of Fig. 2(a). The temperature of the sample measured by an attached RuO_2 thermometer exhibits a minimum after demagnetization. This minimum temperature is $T_{\rm f}$ from $T_{\rm i} = 1.8$ K by ADR. In Fig. 2(b), we plot the $B_{\rm i}$ dependence of $T_{\rm f}$ obtained from our ADR experiments and those evaluated from $S_{\rm el}$ in Fig. 1(b). Here, $T_{\rm f}$ is defined as the temperature at the intersection between $S_{\rm el}(T, B = 0)$ and a line parallel to the horizontal (temperature) axis from $S_{\rm el}(T = T_{\rm i}, B \neq 0)$. These two $T_{\rm f}$ evaluated from $S_{\rm el}$ and the ADR experiments well overlap with each other. $T_{\rm f}$ decreases with increasing $B_{\rm i}$ and reaches 0.26 K at 12 T. The adiabatic temperature change $\Delta T_{\rm ad} = T_{\rm i} - T_{\rm f}$ evaluated from $S_{\rm el}$ is displayed in the inset of Fig. 2(b) as a function of $T_{\rm i}$.

Figure 3(a) and (b) display the magnetic-field dependence of the magnetization M and the field derivative dM/dB down to 0.4 K, respectively. The value of M even above 5 T is $\sim$ 2 $\mu_{\rm B}$/Yb which is about half of 4.00 $\mu_{\rm B}$/Yb expected from the Yb^{3+} free ion without the crystalline-electric-field effect. dM/dB in Fig. 3(b) exhibits a peak due to the metamagnetic transition between 0.9 K and 3 K as shown by the arrows. In the inset of Fig. 3(b), we show the $B-T$ phase diagram constructed with the peak in dM/dB, the drop due to the field-induced phase transition above 2 K, and the peak below 0.5 K in $C_{\rm el}/T$ of Fig. 1(a). This phase boundary of the field-induced phase transition is almost consistent with the reported one[22]. Notably, absence of the peak in dM/dB below 0.5 K suggests this phase boundary reaches the finite temperature in the zero magnetic field.

From two different measurements of specific heat and magnetization, $-\Delta S_{\rm M}$ individually can be evaluated as shown in Fig. 4. Through the Maxwell relation of $(\partial S/\partial B)_T = (\partial M/\partial T)_B$, $-\Delta S_{\rm M}$ as a function of B at a temperature $T = (T_1 + T_2)/2$ is derived from a pair of the isothermal magneti-

zation data at two temperatures of T_1 and T_2 as,

$$-\Delta S_M(B,T) = -\int_0^B \frac{\mathrm{d}M(B',T)}{\mathrm{d}T}\mathrm{d}B' \cong -\int_0^B \frac{M(B',T_1)-M(B',T_2)}{T_1-T_2}\mathrm{d}B'.$$

$-\Delta S_{\mathrm{M}}(T)$ evaluated from the magnetization in Fig. 3(a) is plotted by the open circles in Fig. 4. $S_{\mathrm{el}}(T)$ obtained from the specific heat data in Fig. 1 also provides $-\Delta S_{\mathrm{M}}(B) = S_{\mathrm{el}}(B = 0) - S_{\mathrm{el}}(B)$ as a function of temperature. At 3 T, $-\Delta S_{\mathrm{M}}(T)$ derived from the specific heat and magnetization data exhibits a peak around 1 K, where the value of $-\Delta S_{\mathrm{M}}$ is ~ 0.15 J/K cm^3. This peak goes to the higher temperature side with increasing magnetic fields. For comparison, we plot $-\Delta S_{\mathrm{M}}(T)$ at 3 T in the reported Yb-based metallic magnetic refrigerants to provide sub-Kelvin temperature[13–16,25]. The coexistence of the largest $-\Delta S_{\mathrm{M}}(T)$ of $YbCo_2$ among them and T_{f} below 0.3 K as shown in Fig. 2 indicate the utilities as the cryogenic magnetic refrigerants. As the magnetic refrigerants exhibiting T_{f} below $\sim$ 1 K and evidently exceeding $-\Delta S_{\mathrm{M}} \sim 0.15$ J/K cm^3 around 3 T, some rare-earth oxides/fluorides of the insulators have been reported[3,5,6,8]. Notably, $-\Delta S_{\mathrm{M}} \sim 0.2$ J/K cm^3 has been reported in a Er($4f^{11}$)-based intermetallic compound $ErPd_2Sb$[26].

Materials containing high-density magnetic atoms like $YbCo_2$ are expected to possess a large $-\Delta S_{\mathrm{M}}$ per volume. However, in general, since the strength of the magnetic interaction or magnitude of T_{M} is correlated with the density of magnetic ions, such a large $-\Delta S_{\mathrm{M}}$ competes with the low $T_{\mathrm{M}} \approx T_{\mathrm{f}}$ below 1 K. The field-induced order in $YbCo_2$ is sufficiently suppressed by decreasing the magnetic field to zero, as shown in the inset of Fig. 3(b). In other words, $YbCo_2$ at zero field is located around the boundary between the magnetically ordered phase and the paramagnetic disordered one as the magnetic quantum critical point. This suppression of T_{M} results in the low T_{f} even in the high-Yb-density environment. Indeed, the minimization of T_{f} is observed in the vicinity of the boundary between magnetically ordered phase and paramagnetic disordered one by substituting Cu with Ni in an antiferromagnet Ce_2Cu_2In[19]. In addition, since emergence of field-induced order in $YbCo_2$ suppresses the entropy under the magnetic field $S_{\mathrm{el}}(B \neq 0)$, this field-induced order contributes to increase of $-\Delta S_{\mathrm{M}} = S_{\mathrm{el}}(B = 0) - S_{\mathrm{el}}(B \neq 0)$. Thus, the large $-\Delta S_{\mathrm{M}}(T)$ can coexist with the low T_{f} below $\sim$ 1 K because the ground state at zero field is located around the boundary between the field-induced ordered phase and paramagnetic one.

In conclusion, we report the performance of a Yb-based intermetallic compound $YbCo_2$ as the magnetic refrigerants for cooling down to sub-Kelvin temperature by measuring the specific heat and magnetization, in addition to the ADR experiments. The magnetic entropy change $-\Delta S_{\mathrm{M}}(T)$ per volume exhibits a maximum in $B = 3$ T at $\sim$ 1 K, where the value of $-\Delta S_{\mathrm{M}}$ is ~ 0.15 J/K cm^3. This value is particularly large in the reported Yb-based intermetallic magnetic refrigerants for sub-Kelvin temperature. The large $-\Delta S_{\mathrm{M}}(T)$ and the low T_{f} below 0.3 K are attributed to the ground state, at zero field, locating around the boundary between the field-induced ordered phase and paramagnetic one.

In Supplementary Material, we show the raw data of the specific heat before subtracting the nuclear and lattice contribution.

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AUTHOR DECLARATIONS

CONFLICT OF INTEREST

The authors have no conflicts to disclose.

AUTHOR CONTRIBUTIONS

Yasuyuki Shimura: Conceptualization (lead); Investigation (supporting); Data curation (supporting); Writing - original draft (lead); Writing - review & editing (equal); Supervision (lead). **Ryoma Yokoo:** Investigation (lead); Data curation (lead); Writing - review & editing (equal). **Kanta Watanabe:** Investigation (supporting); Data curation (supporting); Writing - review & editing (equal). **Hiroto Furuie:** Investigation (supporting). **Naohito Tsujii:** Conceptualization (supporting); Writing - review & editing (equal). **Kazunori Umeo:** Investigation (supporting); Writing - review & editing (equal). **Takahiro Onimaru:** Investigation (supporting); Writing - review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available within the article and the supplementary material.

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