Cooling achieved by rotating an anisotropic superconductor in a constant magnetic field: A new perspective

Manh-Huong Phan and David Mandrus

Citation: AIP Advances **6**, 125022 (2016); doi: 10.1063/1.4972124
View online: http://dx.doi.org/10.1063/1.4972124
View Table of Contents: http://aip.scitation.org/toc/adv/6/12
Published by the American Institute of Physics

AIP ADVANCES **6**, 125022 (2016)

Cooling achieved by rotating an anisotropic superconductor in a constant magnetic field: A new perspective

Manh-Huong Phan[1,a] and David Mandrus[2]
[1]*Department of Physics, University of South Florida, Tampa, Florida 33620, USA*
[2]*Department of Materials Science and Engineering, University of Tennessee, Knoxville, Tennessee 37996, USA*

(Received 27 February 2016; accepted 29 November 2016; published online 15 December 2016)

A new type of rotary coolers based on the temperature change (ΔT_{rot}) of an anisotropic superconductor when rotated in a constant magnetic field is proposed. We show that at low temperature the Sommerfeld coefficient $\gamma(B, \Theta)$ of a single crystalline superconductor, such as MgB_2 and NbS_2, sensitively depends on the applied magnetic field (B) and the orientation of the crystal axis (Θ), which is related to the electronic entropy (S_{E}) and temperature (T) via the expression: $S_{\mathrm{E}} = \gamma T$. A simple rotation of the crystal from one axis to one another in a constant magnetic field results in a change in γ and hence S_{E}: $\Delta S_{\mathrm{E}} = \Delta\gamma T$. A temperature change $-\Delta T_{\mathrm{rot}} \sim 0.94$ K from a bath temperature of 2.5 K is achieved by simply rotating the single crystal MgB_2 by 90° with respect to the *c*-axis direction in a fixed field of 2 T. ΔT_{rot} can be tuned by adjusting the strength of B within a wide magnetic field range. Our study paves the way for development of new materials and cryogenic refrigerators that are potentially more energy-efficient, simplified, and compact. [http://dx.doi.org/10.1063/1.4972124]

Refrigeration using solid substances has significant advantages over gas compression/expansion based cooling techniques.[1–3] In particular, magnetic refrigeration based on the temperature change of a magnetic material in response to a magnetic field (the so-called magnetocaloric effect - MCE) has emerged as a promising technology for the development of more energy-efficient and friendly environmentally (chlorofluorocarbon-free) refrigerators.[3–6] Since this technology uses the magnetizing/demagnetizing principle to lower temperature of a magnetocaloric material in a refrigeration cycle,[3] a large mechanical energy is practically required for moving the material in and out of the magnetic field source.[7] In this context, exploitation of the anisotropy of MCE (also known as the *anisotropic* MCE) in anisotropic magnetocaloric materials may offer a good approach to the aforementioned issue.[8–12] Due to the anisotropic property, a large difference in magnetic entropy can be expected when the material is magnetized along the easy and hard magnetization axis directions.[10] In other words, the magnetic entropy change and hence the temperature change of a magnetocaloric material can be realized by simply rotating the material by 90° from the hard magnetization axis to the easy magnetization axis in a fixed magnetic field.[10,11] Keeping the magnetocaloric material within a constant magnetic field enables the conception of rotary magnetic refrigerators working at high frequency, thus leading to the cooling power enhancement and the increased compactness of the devices.[8,11] The giant rotating MCEs have recently been reported in single crystals of $NdCo_5$, $HoMn_2O_5$, and PrSi.[10–12] Continuing efforts are to search for materials with enhanced anisotropic magnetocaloric properties.

An approach to develop solid-state electronic coolers based on tunnel junctions between a normal metal and superconductors for cryogenic refrigeration is also a worthy note.[13–15] This technology relies primarily on the phenomenon that a normal metal - insulator - superconductor junction exhibits

[a]Corresponding author: phanm@usf.edu

electronic cooling in the normal metal, when biased at a voltage just below the superconductor's gap. A refrigerator based on vanadium (V) with a critical temperature of ∼4 K has been reported to efficiently cool down electrons in an Al island from 1 K to 0.4 K.[15] However, this type of refrigerator usually suffers from a poor evaluation of highly energetic quasiparticles in the superconducting electrodes.

In this Letter, we propose a new type of solid-state cooling devices based on the concept of a rotary magnetic refrigerator and anisotropy of the Sommerfeld coefficient of a superconducting material. We show that cooling can be achieved by simply rotating an anisotropic superconductor by 90^o within a constant magnetic field. The potential importance of the proposed device for cryogenic refrigeration is discussed.

According to the thermodynamic theory, the total entropy (S_T) of a material is a sum of the lattice entropy (S_L), electronic entropy (S_E), and magnetic entropy (S_M), which can be expressed as follows:

$$S_T = S_L + S_E + S_M, \tag{1}$$

with S_L being described by[8,9]

$$S_L = -3NR\, ln\left[1 - exp\left(-\frac{T_D}{T}\right)\right] + 12NR\left(\frac{T}{T_D}\right)^3 \int_0^{T_D/T} \frac{x^3}{\exp(x) - 1} dx, \tag{2}$$

S_E being described by

$$S_E = \gamma T, \tag{3}$$

and S_M being described by[16]

$$S_M = NR\left[\sum\nolimits_\sigma \int_{-\infty}^{+\infty} ln\left\{1 + exp\left[-\beta\left(\varepsilon - \mu\right)\right]\right\} \rho_\sigma^d\left(\varepsilon\right) d\varepsilon + \frac{1}{k_B T}\sum\nolimits_\sigma \int_{-\infty}^{+\infty} \left(\varepsilon - \mu\right) f(\varepsilon) \rho_\sigma^d\left(\varepsilon\right) d\varepsilon\right], \tag{4}$$

where N is the number of spins, R the universal gas constant, T_D the Debye temperature, γ the Sommerfeld coefficient, k_B the Boltzmann constant, μ the chemical potential, ε the energy level, $f(\varepsilon)$ the Fermi distribution function, and $\rho_\sigma^d(\varepsilon)$ the density of states. Here we intentionally describe S_M for itinerant electron systems,[16] since type-II superconductors such as MgB_2 and NbS_2, as discussed in this paper, are considered like common metals. From equations (2), (3), and (4), it appears that S_M depends on both magnetic field (B) and temperature (T), while S_L and S_E depend on temperature only.

In most of the previous MCE studies[3,4,6] only the change in magnetic entropy (ΔS_M) with respect to magnetic field was considered in contributing to the total entropy change (ΔS_T). However, we recall that a volume change can affect the phonon system and hence the Debye temperature (T_D) by

$$\frac{\Delta T_D}{T_D} = -\eta\frac{\Delta V}{V}, \tag{5}$$

where η is the Gruneisen parameter, which is between 1 and 3 for many systems.[17] It follows that application of an external magnetic field can cause the volume of the material to change significantly, especially around its structural/magnetic phase transition temperature, leading to the change in T_D and hence ΔS_L, which, in addition to the ΔS_M, contributes to the ΔS_T.[18,19] In this regard, S_L clearly depends not only on temperature, but also on the applied magnetic field. Through direct measurements of the MCE in NiCoMnIn alloys, Kihara *et al.* obtained an extremely large value of ΔS_L (∼51 J/kg K), demonstrating the dominant contribution of ΔS_L to the ΔS_T in this type of material.[19]

On the other hand, the Sommerfeld coefficient (γ) has been reported to depend on both temperature and magnetic field in several magnetic materials.[18,20–22] Since there is a large difference in γ between the antiferromagnetic state and the ferromagnetic one in metamagnetic materials, such as CoMnSi alloys,[18] application of a sufficiently high magnetic field can convert the AFM into the FM phase, resulting in a large change in γ and hence the large ΔS_E via the relation: $\Delta S_E = T\Delta\gamma$. Barcza *et al.* reported $\Delta\gamma \sim 4.7$ mJ/mol K^2 and $\Delta S_E \sim 10$ J/kg K for the CoMnSi alloy, demonstrating a sizable effect of the field-induced variation in γ (which is a measure of the electronic density of states) and hence the dominant contribution of ΔS_E to the ΔS_T in this material.[18] It has also been noted that γ is different with respect to orientation of the material (γ depends on Θ - the angle between the applied magnetic field and the crystal axis), especially for single crystals with strong magnetocrystalline anisotropy.[21,22] As a result, a rotation of the crystal by 90° from the hard magnetization axis to the easy magnetization axis can lead to a change in γ and hence ΔS_E. This follows that in addition to the rotating magnetic entropy change (ΔS_M^{rot}), this rotating electronic entropy change (ΔS_E^{rot}) may also contribute significantly to the total rotating entropy change (ΔS_T^{rot}) of the material,[8–19,21,22] but the prediction of which remains to be verified experimentally.

Based on the strong dependence of the Sommerfeld coefficient (γ) on the magnetic field and the crystal axis (c) in anisotropic superconductors, such as single crystals of MgB_2,[23] $Al_{1-x}Mg_xB_2$,[24] and NbS_2,[25] we have simulated in FIG. 1a a general dependence of γ on B at a given T for $B\,//\,c$ and $B \perp c$. It can be seen in this figure that γ is almost equal for $B \le B_1^{cri}$ and for $B \ge B_2^{cri}$ (B_1^{cri} and B_2^{cri} are the critical magnetic fields, whose magnitudes depend upon materials).[23–25] However, there is a large difference in γ for $B_1^{cri} < B < B_2^{cri}$ (see FIG. 1b). This indicates that a large ΔS_E^{rot} can be realized by simply rotating the crystal by 90° from the perpendicular ($B \perp c$) to parallel ($B\,//\,c$) direction of the c-axis. The corresponding temperature change (ΔT_{rot}) due to this rotation can be estimated as

$$\Delta T_{rot} = -T\frac{\Delta S_E^{rot}}{C_p} = -T^2\frac{\Delta\gamma}{C_p}, \quad (6)$$

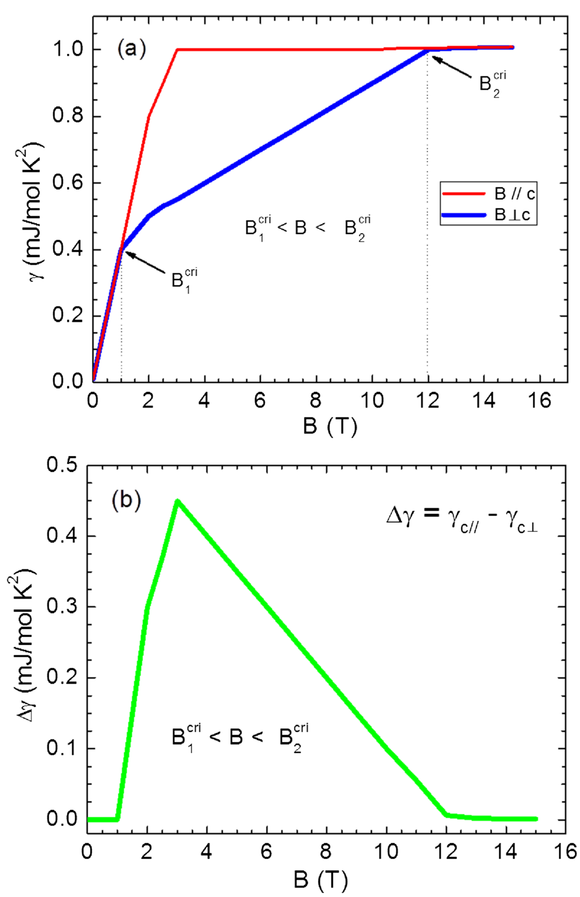

FIG. 1. (**a**) Simulated magnetic field dependence of the Sommerfeld coefficient (γ) of an anisotropic superconductor at a given temperature (T) for the magnetic field parallel ($B\,//\,c$) and perpendicular ($B \perp c$) to the c-axis; (**b**) Simulated magnetic field dependence of $\Delta\gamma$ as the difference in γ between $B\,//\,c$ and $B \perp c$.

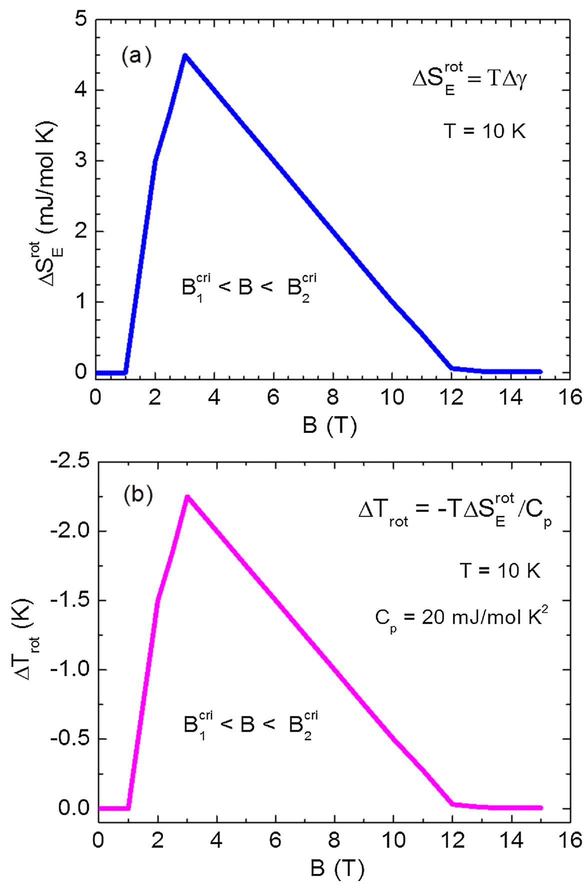

FIG. 2. Simulated magnetic field dependence of the rotating electronic entropy change (ΔS_E^{rot}) and the rotating electronic temperature change (ΔT_{rot}) at a given temperature (T) for an anisotropic superconductor.

where C_p is the specific heat of the material. From the magnetic field dependence of $\Delta\gamma$ in FIG. 1b and equation (6), and considering C_P to be magnetic field independent in the considered low temperature range, the magnetic field dependences of ΔS_E^{rot} and ΔT_{rot} can be simulated, the results of which are shown in FIG. 2. It can be observed that both ΔS_E^{rot} and ΔT_{rot} reach the maximal values at a certain magnetic field under which the largest difference in γ is obtained. Interestingly, ΔT_{rot} is tunable by adjusting the strength of B, which is beneficial for the design of active refrigerators. Taken $\Delta\gamma \sim 1.5$ mJ/mol K^2 and $C_P \sim 10$ mJ/mol K for MgB_2 for $B = 2$ T and T = 2.5 K from Refs. 23 and 24, we have estimated $\Delta S_E^{rot} \sim 3.75$ mJ/mol K and $-\Delta T_{rot} \sim 0.94$ K. This indicates the capacity of cooling MgB_2 from a bath temperature of 2.5 K to 1.56 K by rotating the crystal by 90^o from the c-axis direction perpendicular to the direction of a constant field of 2 T. Larger values of ΔT_{rot} can be expected at lower temperatures for such materials, as $\Delta\gamma$ tends to increase and C_P is decreased with lowering temperature.[23–25] This is of particular interest in exploiting the proposed technique for ultralow temperature cooling applications. Nevertheless, we must note that in a degenerate electron system like MgB_2,[23] spin is an index indicating electronic states rather than an independent local moment. As a result, in this system the magnetic entropy associated with spins of itinerant electrons near Fermi level cannot be simply separated from the total electronic entropy. Since the density of these spins is relatively small, their response to an applied external magnetic field may be small, possibly contributing a tiny cooling effect. Further experimental studies are needed to verify this.

Based on the above findings, we propose in FIG. 3 two possible ways for the design of new rotary refrigerators and their implementations. The first design is similar to that proposed previously for a rotary magnetic refrigerator,[19] where a refrigerator constituted of the superconducting

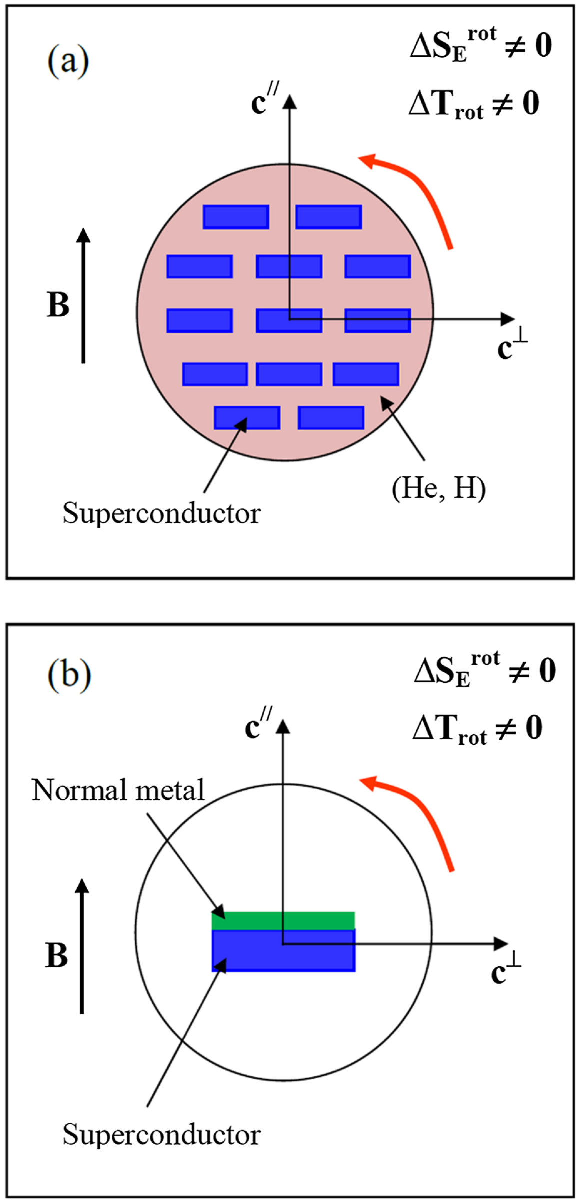

FIG. 3. Proposed cooling applications using superconducting materials as refrigerants and the principle of rotating them in the presence of a constant magnetic field. (a) The first design can be used for the liquefaction of helium and (b) the second design can be used for cooling down a normal metal.

single crystals blocks, instead of using the magnetocaloric single crystals blocks, can be designed (see FIG. 3a). By rotating the crystal axis by 90° from the perpendicular to parallel direction of the *c*-axis, the ΔS_E^{rot} and hence $\Delta T_{\rm rot}$ can be realized. This method is potentially useful for the liquefaction of helium. The second design is based on the cooling principle of a normal metal/superconductor junction, but, instead of using an external voltage to bias the junction near the superconducting gap, a layered film material composed of a normal metal and an anisotropic superconductor can be simply rotated by 90°, allowing the normal metal to be cooled down to a lower temperature due to the cooled superconducting layer (see FIG. 3b). Alternatively, the film is fixed while the magnetic field is rotated by 90°. It has been reported that the sensitivity of ultralow temperature detectors, such as superconducting quantum interference proximity transistor sensors, is significantly improved when the devices are maintained to operate in the sub-50-mK temperature regime.[26] Since the presently proposed technique allows cooling down within the sub-50-mK temperature region by selecting suitable superconducting materials, it potentially opens up new opportunities for applications of a cooling device for advanced microelectromechanical systems (MEMS) and nanoelectromechanical systems (NEMS).

In summary, we have proposed a new approach for the development of rotary coolers using anisotropic superconductors and their various temperature responses with respect to rotating the material axis by 90^{o} in a constant magnetic field. The proposed method may find its usefulness in cryogenic refrigeration technology. Future research should focus on seeking superconducting materials that exhibit large rotating electronic entropy changes in low magnetic field range (< 2 T), in order to exploit this concept fully.

ACKNOWLEDGMENTS

Work at USF was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award # DE-FG02-07ER46438. We thank Dr. Xavier Moya of Cambridge University and Prof. Jenny Hoffman of Colombia University for their valuable discussions and suggestions.

[1] A. Smith, C. R. H. Bahl, R. Bjork, K. Engelbrecht, K. K. Nielsen, and N. Pryds, Adv. Energy Mater. **2**, 1288 (2012).
[2] X. Moya, S. Kar-Narayan, and N. D. Mathur, Nature Materials **13**, 439 (2014).
[3] K. A. Gschneidner, Jr., V. K. Pecharsky, and A. O. Tsokol, Rep. Prog. Phys. **68**, 1479 (2005).
[4] E. Brück, J. Phys. D: Appl. Phys. **38**, R381 (2005).
[5] K. G. Sandeman, Scripta Mater. **67**, 566 (2012).
[6] V. Franco, J. S. Blazquez, B. Ingale, and A. Conde, Annu. Rev. Mater. Res. **42**, 305 (2012).
[7] X. Moya, E. Defay, V. Heine, and N. D. Mathur, Nature Physics **11**, 202 (2015).
[8] P. J. von Ranke, N. A. de Oliveira, E. J. R. Plaza, V. S. R. de Sousa, B. P. Alho, A. M. G. Carvalho, S. Gama, and M. S. Reis, J. Appl. Phys. **104**, 093906 (2008).
[9] E. J. R. Plaza, V. S. R. de Sousa, P. J. von Ranke, A. M. Gomes, D. L. Rocco, J. V. Leitão, and M. S. Reis, J. Appl. Phys. **105**, 013903 (2009).
[10] S. A. Nikitin, K. P. Skokov, Yu. S. Koshkid'ko, Yu. G. Pastushenkov, and T. I. Ivanova, Phys. Rev. Lett. **105**, 137205 (2010).
[11] M. Balli, S. Jandl, P. Fournier, and M. M. Gospodinov, Appl. Phys. Lett. **104**, 232402 (2014).
[12] P. K. Das, A. Bhattacharyya, R. Kulkarni, S. K. Dhar, and A. Thamizhavel, Phys. Rev. B **89**, 134418 (2014).
[13] H. Q. Nguyen, M. Meschke, H. Courtois, and J. P. Pekola, Phys. Rev. Applied **2**, 054001 (2014).
[14] M. Camarasa-Gomez, A. di Marco, F. W. J. Hekking, C. B. Winkelmann, H. Courtois, and F. Giazotto, Appl. Phys. Lett. **104**, 192601 (2014).
[15] O. Quaranta, P. Spathis, F. Beltram, and F. Giazotto, Appl. Phys. Lett. **98**, 032501 (2011).
[16] N. A. de Oliveira and P. J. von Ranke, Physics Reports **489**, 89 (2010).
[17] R. E. Taylor, *Thermal Expansion of Solids*, CINDAS Data Series on Materials Properties Vols. 1–4 (ASM International, Materials Park, OH, 1998).
[18] A. Barcza, Z. Gercsi, H. Michor, K. Suzuki, W. Kockelmann, K. S. Knight, and K. G. Sandeman, Phys. Rev. B **87**, 064410 (2013).
[19] T. Kihara, X. Xu, W. Ito, R. Kainuma, and M. Tokunaga, Phys. Rev. B **90**, 214409 (2014).
[20] P. Limelette, H. Muguerra, and S. Hébert, Phys. Rev. B **82**, 035123 (2010).
[21] L. Caron, M. Hudl, V. Höglin, N. H. Dung, C. P. Gomez, M. Sahlberg, E. Brück, Y. Andersson, and P. Nordblad, Phys. Rev. B **88**, 094440 (2013).
[22] M. Hudl, D. Campanini, L. Caron, V. Höglin, M. Sahlberg, P. Nordblad, and A. Rydh, Phys. Rev. B **90**, 144432 (2014).
[23] Z. Pribulova, T. Klein, J. Marcus, C. Marcenat, F. Levy, M. S. Park, H. G. Lee, B. W. Kang, S. I. Lee, S. Tajima, and S. Lee, Phys. Rev. Lett. **98**, 137001 (2007).
[24] Z. Pribulova, T. Klein, J. Marcus, C. Marcenat, M. S. Park, H.-S. Lee, H.-G. Lee, and S.-I. Lee, Phys. Rev. B **76**, 180502 (2007).
[25] J. Kačmarčík, Z. Pribulová, C. Marcenat, T. Klein, P. Rodière, L. Cario, and P. Samuely, Phys. Rev. B **82**, 014518 (2010).
[26] F. Giazotto, J. T. Peltonen, M. Meschke, and J. P. Pekola, Nat. Phys. **6**, 254 (2010).