Comment on "Observation of vortex-lattice melting in $YBa_2Cu_3O_{7-\delta}$ by Seebeck-effect measurements"

M. Ausloos,* H. Bougrine,[†] and M. Houssa[‡]

S.U.P.R.A.S., University of Liège, Institute of Physics B5, Sart Tilman, B-4000 Liege, Belgium

M. Pekala[§]

Department of Chemistry, University of Warsaw, Al. Zwirki i Wigury 101, PL-02-089 Warsaw, Poland

(Received 4 August 1997)

The findings of Ghamlouch *et al.* [Phys. Rev. B **54**, 9070 (1996)] on the jump in the thermoelectric power of high- T_c superconductors below the critical temperature in the presence of large magnetic fields are discussed. The complicated interplay between the vortex lattice thermodynamic transitions and the transport property percolation transitions raises questions on their similarities, differences, and relationships with respect to the (B,T) phase diagram features and the classical Kosterlitz-Thouless transition. A few comments pertain to the experimental details and others on the relevant role of thermomagnetic transport properties. [S0163-1829(99)04102-8]

Ghamlouch et al.¹ have presented some interesting observations of an anomaly in the Seebeck effect on a detwinned $YBa_2Cu_3O_{7-\delta}$ single crystal and attributed the effect to vortex-lattice melting. The temperature dependence of the Seebeck coefficient (S) is expected to closely follow that of the resistivity ρ via the relation $S = (\rho/\rho_n)S_n$, where the subscript n denotes normal-state values. For temperature gradients applied along the b direction, the behaviors of S vs Tand ρ vs T in various magnetic fields B are similar, a sharp rise and knee are found at some temperature T_m , called the melting temperature for the vortex lattice. The rise in S is not as sharp as for the resistivity. Such a jump can be expected to occur because one is also found (i) in the electrical resistivity² and (ii) in the magnetization—the famous Zeldov jump,³ and also in (iii) SQUID magnetometry measurements,⁴ real and imaginary susceptibility,⁵ specific heat,⁶ muon⁷ and neutron⁸ scattering, I-V characteristics⁹ under similar field-temperature conditions. Such drastic features are associated with a first order phase transition for the lattice vortex melting.^{10,11} These results are consistent with computer simulations in the three-dimensional (3D) XY model¹²⁻¹⁶ and others.¹⁷ The first-order-like transition seems to remain even in the presence of some (simulated through the XY exchange integral) static disorder^{14(b)} but not in the presence of point defects (introduced by irradiation).¹⁸

Transport measurements seem to suggest a first order transition, but they cannot provide a definitive proof of such a transition⁹ because they are thought to be sensitive only to moving vortices, and missing those in thermal equilibrium.^{12–16} There is a need therefore to have some further reflection on such findings and their interpretation.

Near the resistivity drop phenomenon, several transitions can be distinguished: (i) the Ginzburg-Landau T_c temperature at which the coherence length of the order parameter *amplitude* diverges, (ii) the percolation transition¹⁹ temperature T_p at which the *phase* of the order parameter is not disturbed anymore by defects or weak links throughout the sample, such that a perfectly coherent conductivity path connects opposite ends of the sample by superconducting subvolumes. That temperature T_p is truly the one at which ρ =0 because the electrical current flows in the system through an "infinite superconducting percolation cluster." An "ordinary" so called XY transition²⁰ is expected. It can also be shown²¹ by using the Bernouilli equation for the superfluid that a jump occurs in the mean kinetic energy and in the electric potential at $T_{\rm KT}$. The T_p temperature can thus be identified with the Kosterlitz & Thouless temperature $T_{\rm KT}$ if a jump is experimentally observed. The predicted exponential jump²² has been used for explaining low field data behavior near the electrical resistivity percolation transition.²³ The percolation temperature seems to have a different value when "measured" from the electrical resistivity, i.e. when $\rho = 0$ or the thermoelectric power, i.e. when S = 0. Both temperature and field dependences could also be different. A question can thus be raised whether the so-called melting line in Fig. 3 of Ghamlouch et al.¹ obtained from TEP data can be immediately compared with the resistivity percolation line for YBCO-123 compounds in an external field.

Therefore it is not obvious that the dS/dT maximum should reveal a (2D or 3D) vortex lattice melting temperature. The above remarks imply that a precise definition of "melting line," "irreversibility line," "glass transition line," "electrical resistivity percolation line" (and maybe some others) should be given along with the "operational comment'' concerning their experimental determination in order to imply some understanding on their sensitivity to the vortex structure evolution. The irreversibility line $T_i(B)$ or $B_i(T)$ separates the (B,T) regions where vortices as a "solid vortex lattice" or a somewhat "disordered vortex glass" are free to move or are frozen. This line might merge with the $T_{\rm KT}$, T_p , T_m , and even T_c lines in some regions as a function of field. The vortex cooperative behaviors should lead to different effects in various properties, including those measuring the scattering of quasiparticles. However, the cooperative behavior in a given thermodynamic phase is not debated

671

upon in Ref. 1 even though the behavior should have a different signature in different physical quantities indeed.

An additional issue is the order of the melting transition as a function of the magnetic field. A smoother behavior (sometimes seen as a first to second order change) is usual in the presence of increasing disorder.^{14(b),24} However, an external parameter such as a magnetic field usually breaks the continuous symmetry change at a second order phase transition and rather turns it into a discrete symmetry change, i.e., to a first order transition²² as defined for equilibrium properties. For the vortex lattice melting transition, the order parameter is the vortex distribution itself, and the vortex might be one of the main sources of dissipation in the transport properties as well. Notice that quasi particle scattering by vortices has to be considered.^{25,26} In fact, the drop in resistivity between liquid and solid (vortex) phases is reduced in the presence of a field because the "solid" is no longer at rest due to the Lorenz force acting on the vortices and on the quasiparticles. Moreover, the nature of the moving state itself may be quite different in the liquid and solid.^{9,14,17} This means that the S(T) jump, if any, should measure the entropy change and the scattering contributions. Thus a jump in a transport property at a precise "ordering" temperature for a system in which a distribution of temperature exists requires a very fine experimental setup and an almost ideal material.

Thus, experimental conditions as those in Ref. 1 should be examined. A source of concern might be about the ac method for TEP measurements, especially when the frequency does not fit the system time constant, because the alternating TEP signals are self cancelled at high enough frequencies. In private correspondence, Aubin has mentioned that the frequency which was used was 1 Hz, and the signal amplitude was dependent on frequency though the $(\Delta V/\Delta T)$ ratio used to measure S(T) was frequency independent. Previously this technique has led to anomalous peaks in S(T)just above the critical temperature—peaks which were somewhat presented as due to intrinsic effects, though this is debatable.

Another concern with the data arises from an amazing value, i.e., the Landau-Ginzburg critical temperature is reported to be equal to 93.6 K for the zero field case. Even

*Electronic address: ausloos@gw.unipc.ulg.ac.be

- [†]Also at Institut d'Electricité Monetefiore B28, Sart Tilman, B-4000 Liege, Belgium. Electronic address: hassan @montefiore.ulg.ac.be
- [‡]Present address: IMEC, Kapeldreef 75, B-3001 Leuven, Belgium. [§]Electronic address: pekala@chem.uw.edu.pl
- ¹H. Ghamlouch, M. Aubin, R. Gagnon, and L. Taillefer, Phys. Rev. B **54**, 9070 (1996).
- ²W. K. Kwok, J. Fendrich, S. Fleshker, U. Welp, J. Downey, and G. W. Crabtree, Phys. Rev. Lett. **72**, 1092 (1994).
- ³E. Zeldov, D. Majer, M. Konczykowski, V. B. Geshkenbein, V. M. Vinokur, and H. Shrikman, Nature (London) **375**, 373 (1995).
- ⁴U. Welp, J. A. Fendrich, W. K. Kwok, G. W. Crabtree, and B. W. Veal, Phys. Rev. Lett. **76**, 4809 (1996).
- ⁵T. Ishida, K. Okuda, H. Asaoka, Y. Kazumata, K. Noda, and H. Takei, J. Low Temp. Phys. **105**, 1171 (1996).

assuming a high Y123 oxygenation level, we cannot expect such as a high T_c . Moreover the TEP should be negative at high oxygen content—in contrast to the values given in the paper discussed here. In a private correspondence, Aubin pointed out that the reported data in Ref. 1 is the TEP "absolute value"; this should be emphasized in order to avoid any further misleading interpretation. This positive sign related to a high- T_c value might at first sight suggests a systematic error in temperature determination. This temperature calibration effect is not so important *a priori* for the physics of the specific problem, except for the discussed relationship between transition temperatures measured in *different experiments*.

In fact, such features should be better examined with respect to the other magnetotransport effects such as the Nernst effect and the electrothermal conductivity. Notice that the latter is a very powerful test since the resistivity and thermoelectric power can be *simultaneously* measured on the same sample.²⁷ Unfortunately we have not been able to pinpoint a feature indicating some change of line positions or any shift between the melting line and the resistivity percolation line as in Ghamlouch *et al.*¹ for Y123. Only changes in slopes can be noticed even in thorough investigations. Sometimes some sharp drop (going from high temperature) is seen and discussed in terms of activation energy.^{28,29} The observed jump in TEP has never been reported by any other author we know of, previously including the Sherbrooke group.

We conclude that the jump in TEP should be rechecked with respect to other transport properties,³⁰ and to attribute the jump to a vortex lattice melting might need some further theoretical work taking into account both vortex motion dissipation and quasiparticle scattering.^{25,26} Much work is still needed in order to point out how where the percolation, melting and irreversibility lines exist, their field dependence, and why a first or second order melting transition is expected.

We thank M. Aubin for correspondence on the subject. This joint work was supported by the NATO expert visit program HTECH.EV 970414. This work is part of an ARC (94-99/174) grant from the Ministery of Higher Education through the Research Council of the University of Liège. This work was supported by a KBN 3T09A 00811 grant.

- ⁶A. Schilling, R. A. Fisher, N. E. Phillips, U. Welp, D. Dasgupta, W. K. Kwok, and G. W. Crabtree, Nature (London) **382**, 791 (1996); A. Schilling, R. A. Fisher, N. E. Phillips, U. Welp, W. K. Kwok, and G. W. Crabtree, Phys. Rev. Lett. **78**, 4833 (1997).
- ⁷S. L. Lee, P. Zimmermann, H. Keller, M. Warden, I. M. Savic, R. Schauwecker, D. Zech, R. Cubitt, E. M. Morgan, P. H. Kes, T. W. Li, A. A. Menovsky, and Z. Tarnawski, Phys. Rev. Lett. **71**, 3862 (1993).
- ⁸R. Cubitt *et al.*, Nature (London) **365**, 407 (1993).
- ⁹J. A. Fendrich, U. Welp, W. K. Kwok, A. E. Koshelev, G. W. Crabtree, and B. W. Veal, Phys. Rev. Lett. **77**, 2073 (1996).
- ¹⁰G. W. Crabtree, W. K. Kwok, U. Welp, J. A. Fendrich, and B. W Veal, J. Low Temp. Phys. **105**, 1073 (1996).
- ¹¹D. J. C. Jackson and M. P. Das, Supercond. Sci. Technol. 9, 713 (1996).
- ¹²R. E. Hetzel, A. Sudbø, and D. A. Huse, Phys. Rev. Lett. 69, 518 (1992).

- ¹³Y. H. Li and S. Teitel, Phys. Rev. B 47, 359 (1993); 49, 4136 (1994); T. Chen and S. Teitel, Phys. Rev. Lett. 74, 2792 (1995).
- ¹⁴(a) D. Dominguez, N. Grønbech-Jensen, and A. R. Bishop, Phys. Rev. Lett. **75**, 4670 (1995); (b) **78**, 2644 (1997).
- ¹⁵E. A. Jagla and C. A. Balseiro, Phys. Rev. Lett. **77**, 1588 (1996);
 Phys. Rev. B **55**, 3192 (1997).
- ¹⁶A. K. Nguyen, A. Sudbø, and R. E. Hetzel., Phys. Rev. Lett. 77, 1592 (1996).
- ¹⁷J. A. Fendrich, W. K. Kwok, J. Giapintzakis, C. J. van der Beek, V. M. Vinokur, S. Fleshler, U. Welp, H. K. Viswanathan, and G. W. Crabtree, Phys. Rev. Lett. **74**, 1210 (1995).
- ¹⁸H. Safar, P. L. Gammel, D. A. Huse, D. J. Bishop, W. C. Lee, J. Giapintzakis, and D. M. Ginsberg, Phys. Rev. Lett. **70**, 3800 (1994).
- ¹⁹J. Halbriter, Phys. Rev. B 48, 9735 (1993).
- ²⁰J. M. Kosterlitz and D. J. Thouless, in *Progress in Low Temperature Physics*, edited by D. F. Brewer (North Holland, Amsterdam 1978), Vol. VII B p. 371.
- ²¹O. V. Dimitrova (unpublished).
- ²²P. M. Chaikin and T. C. Lubensky, Principle of Condensed Mat-

ter Physics (Cambridge University Press, Cambridge 1995).

- ²³ Yu Fen Guo, P. H. Duvigneaud, H. Bougrine, and M. Ausloos, Physica C 235-240, 3125 (1994); M. Ausloos, H. Bougrine, P. H. Duvigneaud, and Yu Fen Guo, *ibid.* 251, 337 (1995).
- ²⁴G. Blatter, M. V. Feigel'man, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Rev. Mod. Phys. 66, 1125 (1994).
- ²⁵ V. V. Gridin, S. Sergeenkov, and M. Ausloos, Solid State Commun. 98, 623 (1996).
- ²⁶S. A. Sergeenkov, V. V. Gridin, and M. Ausloos, Z. Phys. B 101, 565 (1996).
- ²⁷ M. Houssa, M. Ausloos, and M. Pekala, Phys. Rev. B 54, R 12 713 (1996).
- ²⁸R. P. Huebener, Supercond. Sci. Technol. 8, 189 (1995).
- ²⁹R. P. Huebener, *Magnetic Flux Structures in Superconductors* (Springer-Verlag, Berlin, 1979).
- ³⁰M. Pekala and M. Ausloos, in *Vortex Lattice Melting Probed By Nernst Effect*, Proceedings of the NATO ASI "High Temperature Superconductors IV," edited by S. M. Bose and R. Kossowsky (Kluwer, Dordrecht, 1999).