Nonequilibrium Second-Order Phase Transition in a Cooper-Pair Insulator

A. Doron,[*] I. Tamir, S. Mitra, G. Zeltzer, M. Ovadia,[†] and D. Shahar
Department of Condensed Matter Physics, The Weizmann Institute of Science, Rehovot 76100, Israel

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In certain disordered superconductors, upon increasing the magnetic field, superconductivity terminates with a direct transition into an insulating phase. This phase is comprised of localized Cooper pairs and is termed a Cooper-pair insulator. The current-voltage characteristics measured in this insulating phase are highly nonlinear and, at low temperatures, exhibit abrupt current jumps. Increasing the temperature diminishes the jumps until the current-voltage characteristics become continuous. We show that a direct correspondence exists between our system and systems that undergo an equilibrium, second-order, phase transition. We illustrate this correspondence by comparing our results to the van der Waals equation of state for the liquid-gas mixture. We use the similarities to identify a critical point where an out of equilibrium second-order-like phase transition occurs in our system. Approaching the critical point, we find a power-law behavior with critical exponents that characterizes the transition.

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Highly disordered superconductors exhibit a superconductor to insulator (SIT) [1,2] quantum phase transition [3] upon varying some parameter of the system, such as the magnetic field (B) [4–6], level of disorder [7], film thickness [8] or charge density [9]. In the B-driven insulating phase, above the critical B (B_c), a highly resistive state emerges [5,10–14]. Both theoretical [15,16] and experimental studies [11,13,17] indicate that this insulator is comprised of Cooper pairs that persist beyond the superconducting phase and become spatially localized.

In this Cooper-pair insulator (CPI), at low temperatures ($T \leq 200$ mK), the current voltage characteristics (I-V's) exhibit an abrupt current jump (ΔI) as the voltage is increased beyond a threshold V (V_{th}) [17] (Fig. 1). Reducing V results in a similar discontinuous drop in I at $V \leq V_{\mathrm{th}}$. ΔI can exceed 5 orders of magnitude.

These discontinuities were initially attributed to the emergence of a new "superinsulating" ground state [18], but later Altshuler *et al.* showed that they can be accounted for by a more mundane physical mechanism involving electron heating [19]. (Such discontinuities are not unique to CPIs: similar results were observed, i.e., in amorphous $\mathrm{Y}_x\mathrm{Si}_{1-x}$ films that are not supercondcuting [20]).

At the heart of this electron-heating framework is the assumption that the system is driven out of thermodynamic equilibrium by the applied V. Deviations from linear I-V result from an increase in electron temperature (T_{el}) above the phonon temperature T_{ph}. A steady-state T_{el} is determined by the heat balance between the experimentally applied joule heating, IV, and the cooling of the electrons via their coupling to the phonons. Observing that the electron-phonon coupling drops rapidly at low T [21,22] and that the resistance (R) obeys an exponential T dependence typical of insulators, Altshuler *et al.* numerically solved the heat-balance equation and found that, below a certain T_{ph}, there are two stable solutions for T_{el}. The current jumps are a result of the electrons abruptly changing their T from the low T_{el} solution (T_{el}^L), where a high R (HR) state exists, to the high T_{el} solution (T_{el}^H) where the electrons have a lower R (LR).

Determining T_{el} for each applied V is achieved by combining equilibrium $R(T)$ measurements with nonequilibrium I-V's. At low V ($V \ll V_{\mathrm{th}}$), the electrons are in thermal equilibrium with the phonons ($T_{\mathrm{el}} = T_{\mathrm{ph}}$). Therefore, low-$V$ $R(T)$ measurements can be used as an "electron thermometer," associating a well-defined T_{el} to

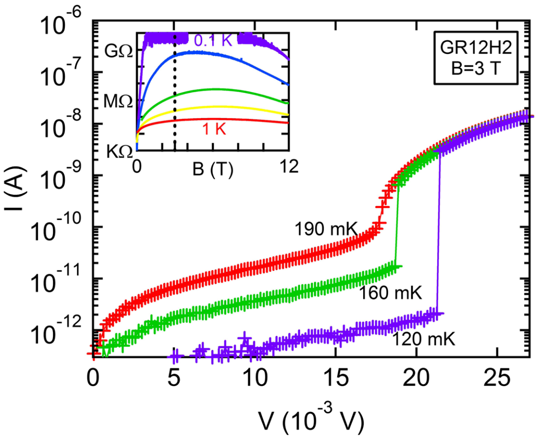

FIG. 1. Discontinuity in the I-V characteristics. I (using a logarithmic scale) vs V. The data points are marked by crosses and the solid lines are a guide to the eye. At $T = 120$ mK, a ΔI of 3 orders of magnitude is apparent at $V_{\mathrm{th}} = 21$ mV. ΔI decreases with T and the I-V turns continuous at $T \geq 190$ mK. Inset: R (logarithmic scale) vs B. The color-coded T's are (from red to purple) $T = 1$, 0.6, 0.4, 0.2, and 0.1 K. The dashed black line marks $B = 3$ T, where the I-V's in the main figure were measured.

each value of R. Since the electron-heating framework relates any deviation from Ohmic I-V's with an increase in T_{el}, for all V's, $V/I = R(T_{\mathrm{el}})$ and the $I(V)$'s can be converted to $T_{\mathrm{el}}(V)$ (a detailed description of this procedure can be found in Ref. [23]). This theoretical approach obtained good support from experiments [23,24].

Altshuler *et al.* also noticed that the discontinuities in T_{el} vs V (T_{el}-V's) are reminiscent of a thermodynamic system undergoing a first-order phase transition. The HR to LR transition can proceed, they stressed, via different metastable paths. This leads to a rich set of phenomena that is expected near V_{th}, a sample of which we have reported on in Ref [25].

In this Letter we follow the evolution of the discontinuity in T_{el}-V as a function of T_{ph}. We find that $\Delta T_{\mathrm{el}} = T_{\mathrm{el}}^{H} - T_{\mathrm{el}}^{L}$ decreases with T_{ph}, and vanishes continuously at a well-defined T (T_c). Our main result is that ΔT_{el}'s approach to zero, as $T_{\mathrm{ph}} \to T_c$, proceeds in a similar way to first-order transitions in systems that undergo an *equilibrium* phase transition. We stress this analogy and its consequences regarding our system by comparing it to the van der Walls (vdW) mean-field description of a liquid-gas transition. We emphasize that vdW is not unique and is merely used as an example, and a similar comparison could be repeated using systems of a verity of universality classes.

Our data were obtained from sample GR12H2, a thin film of amorphous indium-oxide. The film was deposited by e-gun evaporation of high purity $\mathrm{In_2O_3}$ onto a $\mathrm{SiO_2}$ substrate in an $\mathrm{O_2}$ rich environment. A Hall-bar pattern with dimensions of $200 \times 100\ \mu\mathrm{m}^2$ and thickness of 30 nm was patterned via standard photolithography techniques onto Au leads. The sample was superconducting at $B = 0$ and became insulating at $B_c = 0.0275$ T (see inset of Fig. 1). The data presented in all the figures [except for Fig. 2(b)] were measured at $B = 3$ T, in the insulating phase.

In Fig. 2(a) we plot the T_{el}-V's isotherms we obtained at T_{ph} ranging from 100–200 mK. The $I(V)$ data were first translated into $T_{\mathrm{el}}(V)$ and then rotated by 90 degrees relative to Fig. 1. The dashed gray line indicates the region in the T_{el}-V plane where the ΔT_{el}'s are observed.

The choice of using a $(-V)$ axis with negative values was done to illustrate the phenomenological similarity we observed between our data and the solution to the vdW equation of state describing a liquid-gas mixture at thermodynamic equilibrium.

vdW's equation, in its normalized version [26], assumes the form of Eq. (1) where $\hat{P} = P/P_c$, $\hat{v} = v/v_c$, $\hat{\tau} = T/T_c$, P is the pressure, v is the molar volume, and P_c, v_c, and T_c are the critical pressure, molar volume, and temperature, respectively.

$$\hat{P} = \frac{8\hat{\tau}}{3\hat{v} - 1} - \frac{3}{\hat{v}^2} \tag{1}$$

A representation of this equation, including Maxwell's construction for the unstable region is shown in Fig. 2(b).

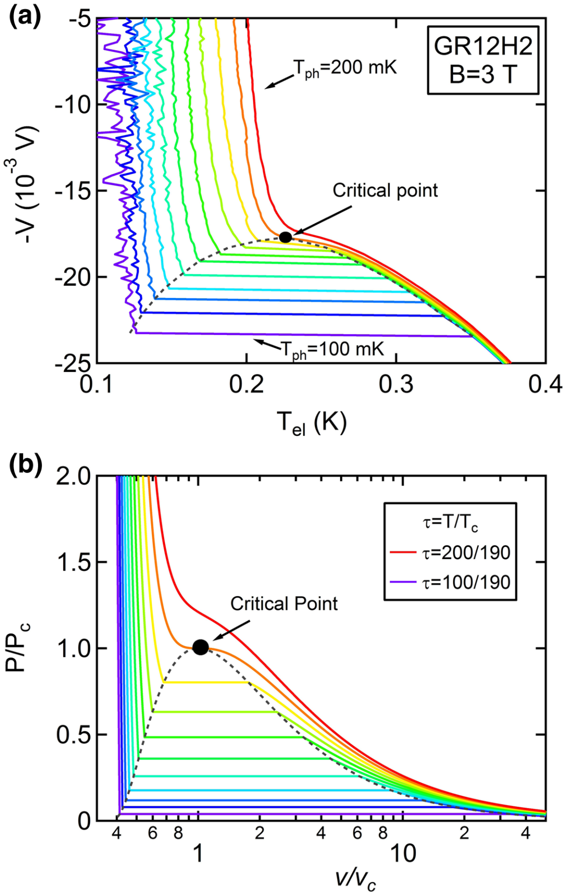

FIG. 2. Resemblance between vdW and T_{el}-V's isotherms. (a) $(-V)$ vs T_{el} isotherms of amorphous indium-oxide in the insulating phase bordering superconductivity. The vdW equivalent critical point is marked by a black circle. The T_{el}-V's turn continuous at $T_{\mathrm{ph}} = T_c = 190$ mK. The color-coded T_{ph}'s are evenly separated by $\Delta T_{\mathrm{ph}} = 10$ mK starting from $T_{\mathrm{ph}} = 100$ mK (purple) up to $T_{\mathrm{ph}} = 200$ mK (red). (b) Solutions of the normalized vdW Eq. (1) with Maxwell's construction on a $\hat{P} - \hat{v}$ diagram (see text). For proper comparison between isotherms of both figures, the value of each τ in (b) equals T_{ph}/T_c of its corresponding color in (a).

The dashed gray line indicates the liquid-gas coexistence region.

A qualitative comparison between 2(a) and 2(b) reveals that both systems share several features: (1) At low T, both systems exhibit large discontinuities. (2) The discontinuities in both systems are largest at the lowest T and decrease as $T \to T_c$. (3) Both systems have a well-defined critical T ($T_c = 190$ mK in the T_{el}-V's and $\tau = 1$ in the $\hat{P} - \hat{v}$ diagram) above which isotherms become continuous.

These similarities imply the existence of an underlying relation, which we explore by constructing a mapping between the two systems, associating $\hat{P}$ and $\hat{v}$ of the gas-liquid mixture with $(-V)$ and T_{el} of the overheated electrons.

In the liquid-gas mixture, the first-order phase transitions terminate at a critical point where a continuous, second-order, phase transition takes place [black circle in Fig. 2(b)]. The central finding of our work is that the unexpected phenomenological correspondence between our $T_{\rm el}$-V's isotherms and the $P-v$ isotherms of the liquid-gas mixture enables us to associate the vanishing of the $\Delta T_{\rm el}$'s upon increasing $T_{\rm ph}$ with the existence of a continuous, second-order like, phase transition. We stress that our system is driven by the large V out of equilibrium conditions. It should be noted that the ΔI's follow similar T evolution as the $\Delta T_{\rm el}$'s; therefore, the pursuing analysis can be repeated for the directly measured I-V's, without making any assumptions regarding the underlying physical nature of the ΔI's in our system (see Supplemental Material [27]).

A central feature of continuous phase transitions is that, approaching the critical point, physical quantities show power law dependences with characteristic critical exponents. To probe the idea of a second-order phase transition in our out of equilibrium CPI, we look for an analogues critical behavior in the vicinity of the critical point. We focus on two critical exponents, β and δ.

In the vdW framework, β characterizes the T evolution of the difference between the gas and liquid v (v_g and v_l) across the jump and is defined by Eq. (2).

$$v_g - v_l \propto (T_c - T)^{\beta} \tag{2}$$

Our system's equivalent critical exponent $\tilde{\beta}$ characterizes the evolution of the $\Delta T_{\rm el}$'s as $T_{\rm ph} \to T_c$ and takes the form of Eq. (3).

$$\Delta T_{\rm el} \propto (T_c - T_{\rm ph})^{\tilde{\beta}} \tag{3}$$

Figure 3(a) displays $\Delta T_{\rm el}$ vs $t_{\rm ph} \equiv (T_c - T_{\rm ph}/T_c)$ on a logarithmic scale. A critical exponent of $\tilde{\beta} = 0.9$ was calculated using a power law fit—dashed black line in the figure. For comparison, in the liquid-gas transition, the theoretical and experimental values of β are 0.5 and 0.34, respectively (the discrepancy is a result of the vdW theory being a mean-field theory).

The critical exponent δ characterizes the change in v, along the critical isotherm, as $P \to P_c$ and is defined by Eq. (4).

$$v_g - v_c \propto |P - P_c|^{1/\delta} \tag{4}$$

The CPI analogous critical exponent, $\tilde{\delta}$, describes the evolution of $T_{\rm el}$ of the LR state along the critical isotherm ($T_{\rm ph} = T_c$) and takes the form of Eq. (5) where V_c and $T_c^{\rm el}$ are the critical V and $T_{\rm el}$.

$$T_{\rm el} - T_c^{\rm el} \propto (V - V_c)^{1/\tilde{\delta}} \tag{5}$$

The blue circles in Fig. 3(b) represent data of $T_{\rm el} - T_c^{\rm el}$ vs $V - V_c$ on a logarithmic scale. Using a power law fit, a critical exponent of $\tilde{\delta} = 1.86$ was calculated. The vdW model predicts $\delta = 3$ and the experimental value is 4.2.

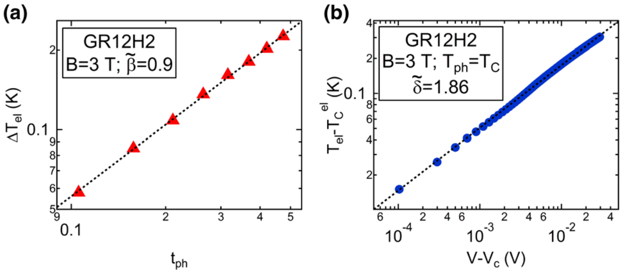

FIG. 3. Electron heating—Critical exponents. (a) $\Delta T_{\rm el}$ vs $t_{\rm ph} \equiv (T_c - T_{\rm ph}/T_c)$ (log-log scale) with $T_c = 190$ mK. The data are presented as red triangles while the power law fit is marked by a dashed black line. The resulting critical exponent is $\tilde{\beta} = 0.9$. (b) $T_{\rm el} - T_c^{\rm el}$ vs $V - V_c$ (log-log scale). The data, in blue circles, were measured along the critical isotherm. The dashed black line is the power law fit. The resulting critical exponent is $\tilde{\delta} = 1.86$.

Discussion.—There are several other phenomena in the liquid-gas transition that find analogues in our system. We list them below.

(1) Coexistence: Using our analogy, by applying a bias V the system is driven out of equilibrium into a steady state where a transition between a HR state of cold electrons and a LR state of hot electrons takes place. Plotting $(-V_{\rm th})$ vs $T_{\rm ph}$ [Fig. 4(a)] yields a curve which is analogous to the coexistence curve in the vdW $P-T$ phase diagram. Measurements that cross this coexistence curve ($T_{\rm ph} < T_c$) result in a discontinuity in $T_{\rm el}$. I-V's measured beyond the critical point ($T_{\rm ph} > T_c$) do not intersect the curve and are continuous. Along the coexistence curve our analogy must imply a phase separation where hot and cold electrons coexist, resulting in (at least) two different values of $T_{\rm el}$ (both of which are higher than $T_{\rm ph}$), and that, close to the critical point, they are intermixed on many length scales. The steady state T difference is maintained by the external power exerted in the I-V measurements.

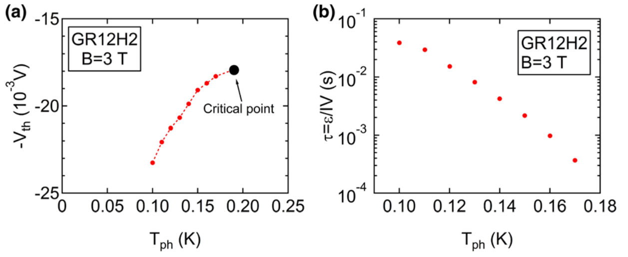

FIG. 4. Phase diagram and heating time. (a) $(-V)$-$T_{\rm ph}$ diagram. The red dots represent $V_{\rm th}$ of each $T_{\rm ph}$ isotherm. This plot is analogous to a vdW $P-T$ phase diagram. The dashed line corresponds to the vapor-liquid coexistence curve and terminates at the critical point. (b) τ vs $T_{\rm ph}$. At low $T_{\rm ph}$ the characteristic heating time is $\tau \sim 10^{-2}$ sec, close to $T_c = 190$ mK $\tau \sim 10^{-4}$ sec.

The coexistence region (the nonaccessible I or T_{el} region) is an unexplored turf. According to the theory (Ref. [19]), the solution of the heat balance equation inside the coexistence region yields an unstable I branch with negative differential resistance. Measuring inside this region is not possible by varying V and measuring I. This is because, as seen in experiments, I discontinuously changes between one of the two values that border the coexistence region. In order to measure inside it one should drive a constant I and measure the V drop across the sample. Conducting this experiment is challenging because it requires a I source that is stable under a load of $\gg G\Omega$ and a voltmeter with internal $R \gg G\Omega$.

(2) Characteristic heating time: The abrupt change in T_{el} of the whole sample is not instantaneous. Its dynamics are considered to be of an avalanchelike process. This process has a characteristic time scale (τ) which is shorter than the time resolution of our measurement (1 sec). In order to roughly assess it we divide the change in electron energy (ϵ), which is found using a free electron approximation, by $P = IV$ at $V = V_{\mathrm{th}}$.

Figure 4(b) displays τ vs T_{ph}, τ ranges between $\tau \sim 10^{-2}$ sec at low T_{ph} and $\tau \sim 10^{-4}$ sec close to the critical point.

(3) Critical slowing down: Outside the coexistence region, external perturbations (such as noise) can drive the system out of its steady-state solution and cause a local transition that will decay with a typical relaxation time. While in equilibrium phase transitions this relaxation time diverges near the critical point, in our case we have not yet been able to test for this possibility.

(4) Universality: Not surprisingly, the critical exponents we extracted do not agree with any known equilibrium universality class. While it is believed that nonequilibrium phase transitions can also be grouped into universality classes, such a classification is still missing. Comparing the critical exponents we obtained to those of 3D directed percolation [28] (a well-studied model that displays a nonequilibrium phase transition) yields a similar $\beta = 0.813$ ($\tilde{\beta} = 0.9$) and a different δ ($\tilde{\delta} = 1.86$).

In summary, we have presented a mapping between a well-known classical system, in thermal equilibrium, and a quantum system, driven far from equilibrium by our I-V measurements. Using this mapping we identified a critical point where a nonequilibrium, second-order, phase transition takes place and found the characteristic critical exponents. Further studies should be made in order to better understand this phase transition and to ascertain what is the dependence of the critical exponents on external parameters such as B, level of disorder, or film thickness.

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[*] Corresponding author.
adam.doron@weizmann.ac.il
[†] Present address: Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA.

[1] A. M. Goldman and N. Markovic, Phys. Today **51**, 39 (1998).
[2] V. F. Gantmakher and V. T. Dolgopolov, Phys. Usp. **53**, 1 (2010).
[3] S. L. Sondhi, S. M. Girvin, J. P. Carini, and D. Shahar, Rev. Mod. Phys. **69**, 315 (1997).
[4] A. F. Hebard and M. A. Paalanen, Phys. Rev. Lett. **65**, 927 (1990).
[5] A. Yazdani and A. Kapitulnik, Phys. Rev. Lett. **74**, 3037 (1995).
[6] T. I. Baturina, D. R. Islamov, J. Bentner, C. Strunk, M. R. Baklanov, and A. Satta, JETP Lett. **79**, 337 (2004).
[7] D. Shahar and Z. Ovadyahu, Phys. Rev. B **46**, 10917 (1992).
[8] D. B. Haviland, Y. Liu, and A. M. Goldman, Phys. Rev. Lett. **62**, 2180 (1989).
[9] K. A. Parendo, K. Tan, A. Bhattacharya, M. Eblen-Zayas, N. E. Staley, and A. M. Goldman, Phys. Rev. Lett. **94**, 197004 (2005).
[10] M. A. Paalanen, A. F. Hebard, and R. R. Ruel, Phys. Rev. Lett. **69**, 1604 (1992).
[11] V. F. Gantmakher, M. V. Golubkov, J. G. S. Lok, and A. K. Geim, JETP **82**, 951 (1996).
[12] G. Sambandamurthy, L. W. Engel, A. Johansson, and D. Shahar, Phys. Rev. Lett. **92**, 107005 (2004).
[13] H. Q. Nguyen, S. M. Hollen, M. D. Stewart, J. Shainline, A. Yin, J. M. Xu, and J. M. Valles, Phys. Rev. Lett. **103**, 157001 (2009).
[14] B. Sacépé, C. Chapelier, T. I. Baturina, V. M. Vinokur, M. R. Baklanov, and M. Sanquer, Phys. Rev. Lett. **101**, 157006 (2008).
[15] M. Feigel'man, L. Ioffe, V. Kravtsov, and E. Cuevas, Ann. Phys. (Amsterdam) **325**, 1390 (2010).
[16] Y. Dubi, Y. Meir, and Y. Avishai, Nature (London) **449**, 876 (2007).
[17] G. Sambandamurthy, L. W. Engel, A. Johansson, E. Peled, and D. Shahar, Phys. Rev. Lett. **94**, 017003 (2005).
[18] V. M. Vinokur, T. I. Baturina, M. V. Fistul, A. Y. Mironov, M. R. Baklanov, and C. Strunk, Nature (London) **452**, 613 (2008).
[19] B. L. Altshuler, V. E. Kravtsov, I. V. Lerner, and I. L. Aleiner, Phys. Rev. Lett. **102**, 176803 (2009).
[20] F. Ladieu, M. Sanquer, and J. P. Bouchaud, Phys. Rev. B **53**, 973 (1996).
[21] M. L. Roukes, M. R. Freeman, R. S. Germain, R. C. Richardson, and M. B. Ketchen, Phys. Rev. Lett. **55**, 422 (1985).
[22] M. E. Gershenson, Y. B. Khavin, D. Reuter, P. Schafmeister, and A. D. Wieck, Phys. Rev. Lett. **85**, 1718 (2000).
[23] M. Ovadia, B. Sacepe, and D. Shahar, Phys. Rev. Lett. **102**, 176802 (2009).
[24] D. Kalok, A. Bilusic, T. I. Baturina, V. M. Vinokur, and C. Strunk, arXiv:1004.5153v1.
[25] O. Cohen, M. Ovadia, and D. Shahar, Phys. Rev. B **84**, 100507(R) (2011).
[26] C. Kittel and H. Kroemer, *Thermal Physics*, 2nd ed. (W. H. Freeman and Company, New York, 1980).
[27] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.116.057001 for a similar analysis for the ΔI's (as opposed to the ΔT_{el}'s) that does not require any assumptions regarding the physical nature of the ΔI's.
[28] S. Lübeck, Int. J. Mod. Phys. B **18**, 3977 (2004).