Parting the Fermi Sea at the Mott Point: Dynamics of Correlated Electrons Reveals the Mechanism Underpinning Mottness

A. Pustogow\textsuperscript{*},\textsuperscript{1,2} R. Rösslhuber\textsuperscript{*},\textsuperscript{1} Y. Tan\textsuperscript{*},\textsuperscript{3} E. Uykur,\textsuperscript{1} A. Böhme,\textsuperscript{1} A. Löhle,\textsuperscript{1} R. Hübner,\textsuperscript{1,4} J. A. Schlueter,\textsuperscript{5,6} M. Dressel,\textsuperscript{1} and V. Dobrosavljević\textsuperscript{3}

\textsuperscript{1}1. Physikalisches Institut, Universität Stuttgart, 70569 Stuttgart, Germany
\textsuperscript{2}Department of Physics and Astronomy, UCLA, Los Angeles, California 90095, U.S.A.
\textsuperscript{3}National High Magnetic Field Laboratory, Florida State University, Tallahassee, U.S.A.
\textsuperscript{4}Institut für Klinische Radiologie und Nuklearmedizin, Universität Heidelberg, Mannheim, Germany
\textsuperscript{5}Material Science Division, Argonne National Laboratory, Argonne, Illinois 60439-4831, U.S.A.
\textsuperscript{6}Division of Materials Research, National Science Foundation, Alexandria, Virginia 22314, U.S.A.
Abstract

By increasing the interaction among conduction electrons, a Fermi-liquid-type metal turns into a Mott insulator. This first-order phase transition should exhibit a regime where the adjacent ground states coexist, leading to electronic phase separation, but the range near $T = 0$ remained unexplored because it is commonly concealed by antiferromagnetism. Here we map the genuine low-temperature Mott transition by applying dielectric spectroscopy under pressure to quantum-spin-liquid compounds. The dielectric permittivity uniquely distinguishes all conduction regimes around the Mott point, allowing us to reliably detect insulator-metal phase coexistence below the critical endpoint. Via state-of-the-art theoretical modeling we establish the coupling between segregated metallic puddles as the driving source of a colossal peak in the permittivity reaching $\epsilon_1 \approx 10^5$ within the coexistence region. Our results indicate that the observed inhomogeneities are the consequence of phase separation emerging from strong correlation effects inherent to Mottness, suggesting a similar 'dielectric catastrophe' in other correlated materials.

MAIN TEXT

Introduction

Many unconventional phenomena and exotic phases are found in the vicinity of the Mott-insulating state, ranging from bad-metal behavior, manifestations of quantum magnetism, to high-temperature superconductivity. This complexity makes it difficult to recognize the dominant degrees of freedom, resulting in a plethora of theoretical viewpoints based on very different ideas. In some cases [1], the anomalies seem associated with symmetry-broken phases – in stark contrast to the early views of Mott and Anderson [2] that spin, charge, or superconducting orders are not the driving force but a consequence of Mottness. The fundamental nature of the Mott point was brought into even sharper focus by the discovery of spin-liquid materials, where geometrical frustration can quench magnetic order down to $T = 0$ [3, 4]. We thus investigated the paradigmatic compound $\kappa$-(BEDT-TTF)$_2$-Cu$_2$(CN)$_3$ by dielectric spectroscopy under pressure (results shown in Fig. 1), which provides a unique opportunity to study the transition from a Mott insulator to a Fermi liquid for 1/2-filled bands in complete absence of magnetic order [4]; not to forget the possibility of unconventional superconductivity stabilized right at the insulator-metal boundary. While
FIG. 1. Dielectric permittivity reveals percolative Mott transition. (A) The genuine Mott state in $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ is characterized by the ac conductivity $\sigma_1(T, p)$ as a function of temperature and pressure. (B) In the permittivity data, a sharply defined insulator-metal phase-coexistence region (sketched on bottom) is identified by the colossal enhancement of $\epsilon_1(T, p)$ (see also 3D plot in the inset). Also shown is the boundary of the Mott insulator, defining the “quantum Widom line” (QWL), and the “Brinkman-Rice line” ($T_{max}$), which delimits the regime of metallic transport, as well as the boundary of the Fermi-liquid regime ($T_{FL}$).

The high-temperature part of the generic phase diagram was widely investigated [5–10], the precise form of the low-temperature transition remains controversial. These studies failed to determine whether the insulator-metal boundary at $T = 0$ should be viewed as a quantum critical point [11, 12], or whether it assumes first-order character (see Fig. 2), as predicted by dynamical mean-field theory (DMFT) [2, 13, 14].

Standard electrical transport is regarded as the decisive experiment for distinguishing
FIG. 2. **Two theoretical scenarios of the Mott insulator-metal transition.** (A) Upon tuning the bandwidth by chemical or physical pressure, for instance, a quasi-continuous transition from a Mott insulator to the Fermi liquid through a quantum critical point at $T = 0$ is suggested for quantum spin liquids with spinon quasiparticles [11]. (B) Dynamical mean-field theory predicts a first-order transition with phase coexistence up to the critical endpoint $T_{\text{crit}}$ [13, 14], and a quantum critical regime associated with the quantum Widom line at higher temperatures [15]. The metallic state is confined by the Brinkman-Rice temperature [16, 17], with the coherent Fermi-liquid regime occurring at lower temperatures.

metals from insulators. Complementary information is commonly extracted from local probes, such as magnetic resonance and optical spectroscopies, etc. Despite considerable effort, these approaches could neither settle the debate about the precise form of the low-temperature Mott insulator-metal transition (IMT), nor prove the proposed phase segregation. Here we discover a strong enhancement of the dielectric permittivity $\epsilon_1(T, p)$ below the critical endpoint of a genuine Mott insulator, shown in Fig. 1, providing unambiguous evidence for percolative phase coexistence around the first-order Mott IMT. We demonstrate that dielectric spectroscopy is superior to these techniques because it (i) allows precise mapping of the phase diagram in dense $(T, p)$ intervals and (ii) is very sensitive to the effects of spatial inhomogeneities on the electrical properties via the complex conductivity. This way we sharply delimit several distinct transport regimes surrounding the Mott point. We substantiate our findings by microscopic modeling of the dielectric response throughout the phase diagram, providing conclusive support for the DMFT-based scenario. This
demonstrates that the dielectric response represents an ideal diagnostic tool to reveal the fundamental physical processes behind Mottness – and insulator-metal transitions in general.

**Results**

For investigating the genuine Mott IMT we select the widely studied compound $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ that is considered to be one of the most convincing quantum-spin-liquid candidates due to its ambient-pressure magnetic properties [3, 18]. We scrutinize the IMT as a function of pressure and temperature by means of dielectric spectroscopy and present our main result in Fig. 1. The ac conductivity $[\sigma_1(T, p) \propto \rho^{-1}(T, p)]$ reproduces previous dc transport results with the transition from the Mott state to a correlated metal featuring Fermi-liquid behavior below $T_{\text{FL}}$ [12, 19]. Likewise, the permittivity $\epsilon_1(T, p)$ distinguishes between the Mott-insulating and metallic regions of the phase diagram, identified as small positive (light red) and large negative (deep blue) values, respectively. The most important feature in Fig. 1B, however, is the regime of extremely large permittivity (bright red) that does not have any counterpart in $\sigma_1$. This prominent peak in $\epsilon_1$ falls exactly on top of the first-order IMT below the critical endpoint at $T_{\text{crit}} \approx 15 – 20$ K [12]. Hence, we assign it to the phase-coexistence region expected from the DMFT scenario (Fig. 2B).

At first glance the result is puzzling because intuitively one expects a smooth, monotonous variation of the permittivity from an insulator with small positive $\epsilon_1^{(i)}$ to large negative values $\epsilon_1^{(m)}$ in a metal due to screening. The situation becomes more delicate in case of a bimodal mixture of phase-segregated metallic and insulating regions – exactly what is anticipated at the first-order Mott IMT (cf. Fig. 2B). As an illustration, consider a toy model of lumped elements, where the pure insulator corresponds to a serial connection of capacitors. As depicted in Fig. 3A,C, introducing conductive puddles essentially “shorts” a finite number of capacitors and, thus, enhances the total capacitance. With larger filling fraction $x$ the resulting $\epsilon_1 \propto C_{\text{total}}$ increases as the growth of metallic islands narrows the distance between the conductive regions and enlarges the cross section of parallel surfaces (highlighted in yellow in Fig. 3D). This capacitive effect leads to a divergence at the percolation threshold $x = x_{\text{crit}}$, which exists strictly only at $T = 0$ (Fig. 3B); a finite peak occurs at finite temperature, as we find (Fig. 1B). Upon further increasing $x > x_{\text{crit}}$ the growing number and size of percolating paths quickly reduces the permittivity from the capacitive to the screening regime ($\epsilon_1 < 0$),
FIG. 3. Capacitive enhancement of permittivity at percolation. (A) Substituting a capacitor ($C_0$) by a resistor ($R_0$) in a serial connection increases the total capacitance $C_2 > C_1$. (B,C) The appearance of metallic puddles (blue) in an insulating medium (red) enhances the macroscopically measured permittivity $\epsilon_1 \propto C_{\text{total}}$ with respect to the insulating value $\epsilon_1^{(i)}$. (D) $\epsilon_1$ diverges when the metallic filling fraction $x$ reaches the percolation threshold ($x = 0.5$ in a 2D system). (E) Strong screening with $\epsilon_1 < 0$ sets in as the metallic state takes over.

and the conductivity of the sample turns metallic (Fig. 3E). Optical spectroscopic results on percolating gold nanoparticle films [20] and near-field nanoimaging on VO$_2$ [21] evidence the capacitive enhancement of $\epsilon_1$ in binary insulator-metal composites.

To further substantiate this physical picture in quantitative details, we carried out theoretical modeling. We calculated the complex dielectric function using DMFT for a single-band Hubbard model, and obtained the respective responses for the insulating and metallic phases around the Mott point [15]. The DMFT phase diagram (Fig. 2B) also features an intermediate coexistence region below $k_B T_{\text{crit}} \sim 0.02W$. In contrast to the homogeneous phases surrounding it, here we expect that even a modest concentration of defects or microstresses induces significant phase separation, leading to percolative effects. To capture it, we postulate smoothly varying volume fractions across the coexistence region (see Methods),
and solve an appropriate electrical network model (cf. Fig. 3A) representing such spatial inhomogeneity.

The real part of the dielectric function in general assumes the form \( \epsilon_1(\omega) = \epsilon_o - \sigma_2(\omega)/\omega \). The second term represents the dynamical contribution of conduction electrons that differs dramatically on the two sides of the Mott IMT. In contrast, the first term corresponds to the (non-universal) dielectric response of the remaining bound electrons in fully occupied bands, that play no significant role at the Mott point. To highlight the effects of Mott localization on the permittivity, we subtract this “background” term and plot \( \epsilon_{1}^{el} = \epsilon_1 - \epsilon_o \) in Fig. 4. The DMFT simulation yields excellent agreement with experiment (Fig. 1) and, most importantly, reproduces in remarkable detail the colossal peak of \( \epsilon_1 \) only within the spatially-inhomogeneous coexistence region.

Our DMFT results further reveal that, in accord with experiment, the permittivity displays remarkably little variation as the transition is approached from the insulator by reducing the Mott gap (see Supplementary Materials for quantitative details). In contrast, the spectacular enhancement of \( \epsilon_1(U/W) \) is sharply confined to the coexistence region, where it exceeds the values of the (homogeneous) Mott insulator by orders of magnitude. This peak appears in an abrupt fashion, very distinct to the smooth and gradual increase in disorder-driven insulator-metal transitions [22]. Physically, it reflects the emergence of a “colossal percolation response” – the hallmark of phase competition in general. Here, it is a smoking gun for strong spatial inhomogeneities arising only within a narrow phase-coexistence region around the Mott point, revealing its fundamental mechanism and content.

**Discussion**

To put our results in proper perspective, we briefly review the available concepts for the bandwidth-driven Mott transition at 1/2-filling. These theories differ significantly in their perspective on what are the dominant physical mechanisms and the related key degrees of freedom in the critical region. Here we focus on those models that disregard (static) spin or charge order as the possible driving force, as appropriate for spin-liquid materials with no symmetry change upon the IMT. One such popular theory [11] puts the emphasis on the long-range spin correlations – “spinons” – contributing to gapless spin excitations in the insulator, and proposes a smooth evolution of the spinon Fermi surface into Fermi-
FIG. 4. DMFT simulations reproduce a peak in $\epsilon_1$. (A,B) $\sigma_1$ and $\epsilon_1$ were calculated in the entire phase diagram by DMFT and the phase-coexistence regime around the first-order IMT. The axes denote the temperature $T$ and Coulomb repulsion $U$ with respect to the bandwidth $W$. (C,D) Fixed-temperature line cuts of the experimental data from Fig. 1 agree well with the simulations plotted in (E,F).

liquid quasiparticles across the transition. The resulting phase diagram (Fig. 2A) displays a certain symmetry with respect to the two phases, predicting a weak temperature dependence of the conductivity in the intermediate (quantum) critical region, while displaying monotonic behavior in the insulator and the metal.

As an alternative viewpoint, DMFT [13] postulates an overwhelmingly local character of the dominant processes with focus on charge excitations, but disregarding the long-range spin correlations. In contrast to the spinon theory, the DMFT phase diagram in Fig. 2B features
not only a first-order transition at $T < T_{\text{crit}}$, but also two distinct crossover lines delimiting dynamical regimes with very different physical character. On the insulating side, the gapped Mott state is bounded by the quantum Widom line [4, 15, 23, 24] with a characteristic “back-bending”; it defines the center of the associated quantum-critical region with a “mirror symmetry” of the resistivity curves [23–25]. On the metallic side, however, DMFT predicts resistivity maxima [16] along the “Brinkman-Rice” line $T_{\text{max}}(p)$, which signal the thermal destruction of resilient quasiparticles [17] and the crossover to semiconducting transport at higher temperatures.

While previous transport and optical studies [4, 24] already provided hints favoring the DMFT scenario, they do not map out the predicted dynamical regimes, especially regarding a well-defined coexistence region at $T < T_{\text{crit}}$. Our new dielectric data, however, reveal all phases in vivid detail and in remarkable agreement with the respective crossover lines obtained from dc transport. Indeed, we recognize the gapped Mott insulator by essentially constant $\epsilon_1$ (light red) bounded precisely by the quantum Widom line [15] (independently determined by transport and optical studies [4]), while also the Brinkman-Rice line [16, 17] clearly follows the dielectric behavior expected for a metal ($\epsilon_1 < 0$, blue). Most remarkably, these two crossover lines converge towards $T_{\text{crit}}$, which marks the onset of the coexistence region, just as anticipated from Fig. 2B. The emergence of phase segregation is evidenced by the huge peak of $\epsilon_1$ in excellent agreement with our current DMFT-based modeling. The sharply defined boundaries of this dielectric anomaly imply that the corresponding inhomogeneities are the consequence and not the cause of phase separation, the latter resulting from strong correlation effects inherent to Mottness.

Our findings leave little room for doubt that the DMFT scenario offers the more accurate picture of the Mott transition region. This also confirms recent experimental and theoretical results [26, 27] suggesting that gapless spin excitations, while dominant deep within the Mott-insulating phase and at very low temperatures, are quickly damped away by the onset of charge fluctuations close to the IMT. In the Supplementary Materials we also show that our DMFT-based theory offers another perspective on the low-temperature resistivity data from Furukawa et al. [12], which was previously interpreted from a different theoretical viewpoint. We explain how phase coexistence has a dramatic effect on the amplitude of the $AT^2$ resistivity term in the conducting phase, which can be directly seen already in the experimental data of Ref. [12]. Our results portray a remarkably simple yet robust picture
of local quantum criticality, a principle with potential relevance and likely consequences much beyond quantum spin liquids at 1/2 filling. From the experimental point of view, our work demonstrates dielectric spectroscopy as an extremely powerful tool, allowing to clearly recognize and precisely delimit various physically distinct regimes around the Mott point. Even a quick look at the color map in Fig. 1 immediately gives an amazing amount of information - perhaps for the first time providing a glimpse into the long-standing puzzle of what goes on around the Mott point.

Our observations call for nm-resolved spectroscopic studies of the complex phase segregation in the coexistence region. Optical near-field microscopy, for instance, is capable of mapping the complex conductivity locally and recent pioneering works reported the first cryogenic experiments on correlated materials [28–30]. However, state-of-the-art nanoimaging is confined to several tens of kelvins due to heating effects caused by the high laser intensity. Future advances may further push the limits of this technology making the low-temperature Mott transition accessible for nanoscopic investigations. While other scanning-probe techniques even reach below 1 K, most of them are sensitive to either metallic or insulating materials, and the requirement to work under high-vacuum renders pressure-tuning impossible. The ability of dielectric spectroscopic experiments to unravel microscopic conduction processes at low-temperatures, along with in situ pressure tuning through the phase transition, puts it to a sweet spot in condensed-matter research, ahead of scanning probes.

Finally, we compare our findings to related materials with a similar dielectric anomaly near an IMT. In $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl the critical endpoint is located at 35 K and 230 bar [6, 31] and Mott physics is similar to the compound studied here [4]. Hence, the coexistence region is likely entered already at ambient pressure when cooling below $T_{\text{crit}}$. Concomitantly, $\epsilon_1$ is strongly enhanced in the same fashion as observed here, suggesting an analogue phase-segregated scenario rather than the previously suggested multiferroicity [32, 33]. Likewise, the “ferroelectric-like” peak at the charge-order IMT of $\kappa$-(BEDT-TTF)$_2$Hg(SCN)$_2$Cl goes hand in hand with insulator-metal coexistence, evident from recent ESR [34] and noise spectroscopy results [35].
METHODS

Experimental. We selected two high quality single crystals of \(\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3\) grown by the standard electrochemical synthesis method [36, 37] at Stuttgart (sample S1) and at Argonne (sample S2). The complex impedance of the crystals was measured as a function of pressure, temperature and frequency to obtain the permittivity \(\hat{\epsilon} = \epsilon_1 + i\epsilon_2\) and the conductivity \(\hat{\sigma} = \sigma_1 + i\sigma_2\). The crystals are contacted out-of-plane (\(E \parallel a\)) by attaching thin gold wires with carbon paint to the opposite crystal surfaces. The measurements were performed with two contacts in a pseudo four-point configuration using an Agilent 4294 Impedance Analyzer. The applied ac voltage was set to 0.5 V which was still in the Ohmic regime. We used a piston-type pressure cell ranging up to 10 kbar with a self-made electrical feedthrough for coaxial cables designed for pressure-dependent dielectric spectroscopy measurements as described in detail elsewhere [38]. Daphne oil 7373 was used as liquid pressure-transmitting medium because it is inert to molecular solids, has a good hydrostaticity and stays fluid at room temperature for all applied pressures. The inherent pressure loss upon cooling was recorded continuously in-situ by an InSb semiconductor pressure gauge [39], which shows a negligible pressure gradient below 50 K. The main temperature range discussed here is collected from the same pressure cycles; and the pressure reading at 10 K is given throughout the manuscript, unless stated otherwise. A continuous flow cryostat was utilized to cool down the pressure cell. A total cable length of only 50 cm enables measurements up to 5 MHz. The rather steep thermal gradient limits the lowest reachable temperature to 8 K. No dependence on the cooling rate was observed, which was kept below 0.4 K/min for all measurements. There is good agreement between the results obtained on both samples and we present data of sample S2 for which we measured more temperature sweeps under varying applied pressures. The results obtained on sample S1, along with additional data on sample S2 including different frequencies, can be found in the Supplementary Materials.

Dynamical Mean-Field Theory Modeling. We performed standard [13, 17] DMFT calculations of the optical conductivity \(\sigma_1\) of a single-band Hubbard model at 1/2-filling, using a semicircular model density of states and as an impurity solver, which suffices [15, 23] for our purposes. The imaginary part of conductivity \(\sigma_2\) was then obtained via the Kramers–Kronig transformation, and the dielectric function \(\epsilon(\omega) = 1 + \frac{i\omega}{\omega}\) was obtained across the entire phase diagram, for both uniform phases. To describe percolative effects within the phase
coexistence region, we use the standard effective medium theory [40] for the corresponding random conductor/dielectric network, with a binary distribution of the local dielectric functions as calculated from DMFT for given temperature $T$ and Coulomb repulsion $U$. All energies are expressed in units of the bandwidth $W$, i.e. $T/W$ and $U/W$. For simplicity, we assign a smooth hyperbolic tangent function to model the volume fraction of the metallic phase: 

$$x(T/W, U/W) = \frac{1}{2} \tanh \left\{ \frac{a}{((T/W)_{\text{crit}} - T/W)\left[ (U/W)_{\text{crit}} - U/W \right]} + \frac{1}{2} \right\},$$

which is centered around the middle of the coexistence region: $(U/W)_{\text{crit}} = (0.20 - T/W)/0.14$; we also selected $a = 0.1$ for $U/W > (U/W)_{\text{crit}}$ and $a = 0.3$ for $U/W < (U/W)_{\text{crit}}$ as expected from the DMFT phase diagram [15, 23].

[1] Masatoshi Imada, Atsushi Fujimori, and Yoshinori Tokura, “Metal-insulator transitions,” Rev. Mod. Phys. 70, 1039–1263 (1998).
[2] N. F. Mott, Metal-Insulator Transitions, 2nd ed. (Taylor & Francis Ltd., Bristol, 1990).
[3] Leon Balents, “Spin liquids in frustrated magnets.” Nature 464, 199–208 (2010).
[4] A Pustogow, M Bories, A Löhle, R Rösslhuber, E Zhukova, B Gorshunov, S Tomić, J A Schlueter, R Hübner, T Hiramatsu, Y Yoshida, G Saito, R Kato, T.-H. Lee, V Dobrosavljević, S Fratini, and M Dressel, “Quantum spin liquids unveil the genuine Mott state,” Nat. Mater. 17, 773–777 (2018).
[5] C N Berglund and H J Guggenheim, “Electronic Properties of VO$_2$ near the Semiconductor-Metal Transition,” Phys. Rev. 185, 1022–1033 (1969).
[6] P. Limelette, A. Georges, D. Jérôme, P. Wietek, P. Metcalf, and J. M. Honig, “Universality and Critical Behavior at the Mott Transition,” Science 302, 89 – 92 (2003).
[7] P Hansmann, A Toschi, G Sangiovanni, T Saha-Dasgupta, S Lupi, M Marsi, and K Held, “MottHubbard transition in V2O3 revisited,” Phys. Status Solidi B 250, 1251–1264 (2013).
[8] P Limelette, P Wietek, S Florens, A Georges, T A Costi, C Pasquier, D Jérôme, C Mézière, and P Batail, “Mott Transition and Transport Crossovers in the Organic Compound $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl,” Phys. Rev. Lett. 91, 16401 (2003).
[9] F Kagawa, K Miyagawa, and K Kanoda, “Unconventional critical behaviour in a quasi-two-dimensional organic conductor,” Nature 436, 534 (2005).
[10] F. Kagawa, T. Itou, K. Miyagawa, and K. Kanoda, “Transport criticality of the first-order Mott transition in the quasi-two-dimensional organic conductor $\kappa-(BEDT-TTF)_2Cu[N(CN)_2]Cl,” Phys. Rev. B 69, 64511 (2004).

[11] T. Senthil, “Theory of a continuous Mott transition in two dimensions,” Phys. Rev. B 78, 45109 (2008).

[12] Tetsuya Furukawa, Kazuhiko Kobashi, Yosuke Kurosaki, Kazuya Miyagawa, and Kazushi Kanoda, “Quasi-continuous transition from a Fermi liquid to a spin liquid in $\kappa$-(ET)$_2$Cu$_2$(CN)$_3$,” Nat. Commun. 9, 307 (2018).

[13] Antoine Georges, Gabriel Kotliar, Werner Krauth, and Marcelo J. Rozenberg, “Dynamical mean-field theory of strongly correlated fermion systems and the limit of infinite dimensions,” Rev. Mod. Phys. 68, 13–125 (1996).

[14] D. Vollhardt, “Dynamical mean-field theory for correlated electrons,” Ann. Phys. (Berl.) 524, 1–19 (2012).

[15] J. Vučičević, H. Terletska, D. Tanasković, and V. Dobrosavljević, “Finite-temperature crossover and the quantum Widom line near the Mott transition,” Phys. Rev. B 88, 75143 (2013).

[16] M. M. Radonjić, D. Tanasković, V. Dobrosavljević, K. Haule, and G. Kotliar, “Wigner-Mott scaling of transport near the two-dimensional metal-insulator transition,” Phys. Rev. B 85, 85133 (2012).

[17] Xiaoyu Deng, Jernej Mravlje, Rok Žitko, Michel Ferrero, Gabriel Kotliar, and Antoine Georges, “How Bad Metals Turn Good: Spectroscopic Signatures of Resilient Quasiparticles,” Phys. Rev. Lett. 110, 86401 (2013).

[18] Y. Shimizu, K. Miyagawa, K. Kanoda, M. Maesato, and G. Saito, “Spin Liquid State in an Organic Mott Insulator with a Triangular Lattice,” Phys. Rev. Lett. 91, 107001 (2003).

[19] Y. Kurosaki, Y. Shimizu, K. Miyagawa, K. Kanoda, and G. Saito, “Mott Transition from a Spin Liquid to a Fermi Liquid in the Spin-Frustrated Organic Conductor $\kappa-(ET)_2$Cu$_2$(CN)$_3$, ” Phys. Rev. Lett. 95, 177001 (2005).

[20] Martin Hövel, Bruno Gompf, and Martin Dressel, “Dielectric properties of ultrathin metal films around the percolation threshold,” Phys. Rev. B 81, 35402 (2010).

[21] M. M. Qazilbash, M. Brehm, Byung-Gyu Chae, P.-C. Ho, G. O. Andreev, Bong-Jun Kim, Sun Jin Yun, A. V. Balatsky, M. B. Maple, F. Keilmann, Hyun-Tak Kim, and D. N. Basov,
“Mott transition in VO\textsubscript{2} revealed by infrared spectroscopy and nano-imaging.” Science 318, 1750–3 (2007).

[22] Jeppe C Dyre and Thomas B Schröder, “Universality of ac conduction in disordered solids,” Rev. Mod. Phys. 72, 873–892 (2000).

[23] H. Terletska, J. Vučičević, D. Tanasković, and V. Dobrosavljević, “Quantum Critical Transport near the Mott Transition,” Phys. Rev. Lett. 107, 26401 (2011).

[24] T. Furu kawa, K. Miyagawa, T. Itō, M. Itó, H. Taniguchi, M. Saito, S. Iguchi, T. Sasaki, and K. Kanoda, “Quantum Spin Liquid Emerging from Antiferromagnetic Order by Introducing Disorder,” Phys. Rev. Lett. 115, 77001 (2015).

[25] V Dobrosavljević, Elihu Abrahams, E Miranda, and Sudip Chakravarty, “Scaling Theory of Two-Dimensional Metal-Insulator Transitions,” Phys. Rev. Lett. 79, 455–458 (1997).

[26] Tsung-Han Lee, Serge Florens, and Vladimir Dobrosavljević, “Fate of Spinons at the Mott Point,” Phys. Rev. Lett. 117, 136601 (2016).

[27] A Pustogow, Y Saito, E Zhukova, B G orshunov, R Kato, T.-H. Lee, S Fratini, V Dobrosavljević, and M Dressel, “Low-Energy Excitations in Quantum Spin Liquids Identified by Optical Spectroscopy,” Phys. Rev. Lett. 121, 056402 (2018).

[28] A. S. McLeod, E. van Heumen, J. G. Ramirez, S. Wang, T. Saerbeck, S. Guenon, M. Goldflam, L. Anderegg, P. Kelly, A. Mueller, M. K. Liu, Ivan K. Schuller, and D. N. Basov, “Nanotextured phase coexistence in the correlated insulator V\textsubscript{2}O\textsubscript{3},” Nat. Phys. 13, 80–86 (2016).

[29] K W Post, A S McLeod, M Hepting, M Bluschke, Yifan Wang, G Cristiani, G Logvenov, A Charnukha, G X Ni, Padma Rad hakrishnan, M Minola, A Pasupathy, A V Boris, E Benckiser, K A Dahmen, E W Carlson, B Keimer, and D N Basov, “Coexisting first- and second-order electronic phase transitions in a correlated oxide,” Nat. Phys. 14, 1056–1061 (2018).

[30] Andrej Pustogow, Alexander S. McLeod, Yohei Saito, Dmitri N. Basov, and Martin Dressel, “Internal Strain Tunes Electronic Correlations on the Nanoscale,” Sci. Adv. 4, eaau9123 (2018).

[31] Elena Gati, Markus Garst, Rudra S Manna, Ulrich Tutsch, Bernd Wolf, Lorenz Bartosch, Harald Schubert, Takahiko Sasaki, John A Schlueter, and Michael Lang, “Breakdown of Hooke’s law of elasticity at the Mott critical endpoint in an organic conductor,” Sci. Adv. 2, e1601646 (2016).

[32] Peter Lunkenheimer, Jens Müller, Stephan Krohns, Florian Schrettle, Alois Loidl, Benedikt Hartmann, Robert Rommel, Mariano de Souza, Chisa Hotta, John A Schlueter, and Michael
Lang, “Multiferroicity in an organic charge-transfer salt that is suggestive of electric-dipole-driven magnetism,” Nat. Mater. 11, 755 (2012).

[33] Marko Pinterić, David Rivas Góngora, Željko Rapljenović, Tomislav Ivek, Matija Čulo, Bojana Korin-Hamzić, Ognjen Milat, Branko Gumhalter, Predrag Lazić, Miriam Sanz Alonso, Weiwu Li, Andrej Pustogow, Guilherme Gorgen Lesseux, Martin Dressel, and Silvia Tomić, “Electrodynamics in Organic Dimer Insulators Close to Mott Critical Point,” Crystals 8, 190 (2018).

[34] Elena Gati, Jonas K. H. Fischer, Peter Lunkenheimer, David Zielke, Sebastian Köhler, Felizitas Kolb, Hans-Albrecht Krug von Nidda, Stephen M Winter, Harald Schubert, John A Schlueter, Harald O Jeschke, Roser Valentí, and Michael Lang, “Evidence for Electronically Driven Ferroelectricity in a Strongly Correlated Dimerized BEDT-TTF Molecular Conductor,” Phys. Rev. Lett. 120, 247601 (2018).

[35] Tatjana Thomas, Benedikt Hartmann, Peter Lunkenheimer, Harald Schubert, John A Schlueter, and Jens Müller, “Low-Frequency Charge Carrier Dynamics in Ferroelectric κ-(BEDT-TTF)2X: A Comparative Study of X = Cu[N(CN)2]Cl and X = Hg(SCN)2Cl,” Phys. Status Solidi B 2019, 1800746 (2019).

[36] Urs. Geiser, Hau H. Wang, K. Douglas. Carlson, Jack M. Williams, Henry A. Charlier, James E. Heindl, George A. Yaconi, Bradley J. Love, Michael W. Lathrop, James E. Schirber, D. L. Overmyer, Jingqing Ren, and Myung-Hwan Whangbo, “Superconductivity at 2.8 K and 1.5 kbar in κ-(BEDT-TTF)2Cu2(CN)3: the first organic superconductor containing a polymeric copper cyanide anion,” Inorg. Chem. 30, 2586–2588 (1991).

[37] Tokutaro Komatsu, Nozomu Matsukawa, Takeharu Inoue, and Gunzi Saito, “Realization of Superconductivity at Ambient Pressure by Band-Filling Control in κ-(BEDT-TTF)2Cu2(CN)3,” J. Phys. Soc. Jpn. 65, 1340–1354 (1996).

[38] R Rösshuber, E Uykur, and M Dressel, “Pressure cell for radio-frequency dielectric measurements at low temperatures,” Rev. Sci. Instrum. 89, 54708 (2018).

[39] R Beyer and M Dressel, “Piston pressure cell for low-temperature infrared investigations,” Rev. Sci. Instrum. 86, 53904 (2015).

[40] Scott Kirkpatrick, “Percolation and Conduction,” Rev. Mod. Phys. 45, 574–588 (1973).
ACKNOWLEDGMENTS

General: We thank G. Untereiner for contacting the crystals and S. Brown for fruitful discussions.

Funding: We acknowledge funding by the Deutsche Forschungsgemeinschaft (DFG) via DR228/52-1. A.P. acknowledges support by the Alexander von Humboldt Foundation through the Feodor Lynen Fellowship. Work in Florida was supported by the NSF Grant No. 1822258, and the National High Magnetic Field Laboratory through the NSF Cooperative Agreement No. 1157490 and the State of Florida. J.A.S. acknowledges support from the Independent Research/Development program while serving at the National Science Foundation.

Author Contributions: Dielectric spectroscopic investigations were conceived by M.D., R.R. designed and performed the experiments, supported by A.B. and E.U.. Y.T. and V.D. carried out theoretical calculations. A.P. initiated and steered the collaboration of experiment and theory. A.L., R.H. and J.A.S. prepared the crystals. A.P., M.D. and V.D. discussed the data, interpreted the results, and wrote the paper with input from all authors. A.P., R.R. and Y.T. contributed equally to this work.

Competing Interests: The authors declare that they have no competing financial interests.

Materials & Correspondence: All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Additional data related to this paper may be requested from the authors. Correspondence and requests for materials and technical details should be addressed to A.P. andrej.pustogow@pi1.physik.uni-stuttgart.de, pustogow@physics.ucla.edu, M.D. dressel@pi1.physik.uni-stuttgart.de or V.D. vlad@magnet.fsu.edu.
A. Determination of phase boundaries from conductivity

The ac conductivity of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ was measured by dielectric spectroscopy, where the real part $\sigma_1(T, p)$ reproduces well the temperature and pressure dependence of previous dc transport results [S1, S2, S3, S4]. In Fig. S1 we show the resistivity $\rho_1 = \sigma_1^{-1}$ acquired at 7.5 kHz, which is closest to the dc limit at $f = 0$. The behavior changes from insulating ($d\rho_1/dT < 0$) to metallic ($d\rho_1/dT > 0$) for increasing pressure, accompanied by a general reduction of resistivity. The boundary of the Fermi liquid, $T_{FL}$, and of the metallic transport regime, associated with the Brinkman-Rice temperature $T_{max}$, were determined by examining $\rho_1(T)$ as illustrated in Fig. S2A,B for exemplary resistivity curves. The maximum in $\rho_1(T)$ straightforwardly gives $T_{max}$ whereas, following the common procedure, $T_{FL}$ is defined as the temperature at which the deviation in $\rho_1(T)$ from the characteristic $T^2$ behavior of a Fermi liquid, $\rho_{1,\text{fit}} = \rho_0 + AT^2$, exceeds 10%. This is most accurately done by plotting $\rho_1$ and $\rho_{1,\text{fit}}$ versus $T^2$. Panel C shows the determination of the

![Graph showing resistivity vs temperature](image)

**FIG. S1.** The temperature-dependent resistivity $\rho_1 = 1/\sigma_1$ is shown for the different applied pressures. These dielectric data of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ acquired at finite frequency ($f = 7.5 \, kHz$) for $E \parallel a$ agree nicely with the previously reported dc resistivity as a function of pressure and temperature [S1, S2, S3, S4].
FIG. S2. (A) \( \rho_1(T) \) of \( \kappa-(BEDT-TTF)_2Cu_2(CN)_3 \) is shown for 1.69 kbar and 7.5 kHz. The location of the maximum yields the Brinkman-Rice temperature \( T_{\text{max}} \). (B) Fermi-liquid behavior is fitted according to \( \rho_{1,\text{fit}} = \rho_0 + AT^2 \) (solid red line) at 1.69 kbar and shown on a quadratic temperature scale together with the experimental data. The red arrow indicates the temperature \( T_{\text{FL}} \) at which \( \rho_1 \) deviates from \( \rho_{1,\text{fit}} \) by more than 10%. (C) For determination of the quantum Widom line the pressure-dependent resistivity was evaluated at constant temperature above the critical endpoint, here exemplarily shown for \( T = 35 \, \text{K} \). The crossover pressure \( p_c \) at the steepest slope is obtained by fitting a tangens hyperbolicus function to the data.

quantum Widom line in the supercritical region \( T > T_{\text{crit}} \); it is defined as the steepest slope of \( \log \rho \) as a function of pressure [S3]. In particular, we extracted the crossover pressure \( p_c \) by fitting a function \( \tanh(p - p_c) \) to the experimental data.

B. Frequency and sample dependence of percolation peak

Dielectric spectroscopic experiments were performed in the range 7.5 kHz – 2.7 MHz, with the best signal to noise ratio between 50 and 700 kHz. The characteristic percolation peak, along with the Mott-insulating and metallic phases, discussed in this work is found at all frequencies. We exemplarily show the \( T - p \) plots of \( \sigma_1 \) and \( \epsilon_1 \) in Fig. S3A-D and E-H, respectively, which each were acquired at 53, 100, 200 and 700 kHz as indicated. Note that the color code for the optical conductivity \( \sigma_1 \) on the right side corresponds to panels A-D and the color code for the permittivity \( \epsilon_1 \) to panels E-H, respectively. While the absolute values of \( \sigma_1 \) are essentially frequency-independent, \( \epsilon_1 \) exhibits a pronounced frequency dependence at the metallic side of the phase diagram (\( p > 2.5 \, \text{kbar}, \, T < T_{\text{max}} \)). In particular, the strength of the percolation peak is strongly reduced with increasing frequency, accompanied by a
less negative screening response in the homogeneous bad-metallic and Fermi-liquid states. Above discussed features are seen best at low temperatures which we illustrate in Fig. S4 showing the pressure dependence at 10 K for different frequencies, i.e. cuts through the phase diagrams in Fig. S3E-H at $T = 10$ K (including lower frequencies and the 380 kHz data shown in Figs. 1 and 4 of the main manuscript). In the incoherent semiconducting regime at elevated temperatures ($T > 100$ K) no dispersion is observed. We further notice that $\epsilon_1$ is more or less frequency-independent in the Mott-insulating state. Here, the background contribution to the permittivity $\epsilon_o$, originating from lattice vibrations and bound electrons in fully occupied bands, prevails upon the low-energy contribution that is determined by the Mott gap $\Delta$. Since at the measured frequency $hf \ll k_B T \ll \Delta$, the slight increase of $\epsilon_1$ with pressure up to 1.2 kbar is associated with the reduction of the Mott gap. This is in full agreement with our DMFT modeling presented below (cf. Fig. S6). Obviously, the abrupt onset and strong increase for $p > 1.2$ kbar clearly corresponds to phase coexistence at the percolative first-order insulator-metal transition (IMT). Although nanoscopic imaging of this region is highly desired, similar optical near-field experiments like those reported on charge-ordered materials [S5, S6] are problematic in the present case because of the required

![FIG. S3](image)

**FIG. S3.** (A-D) $\sigma_1(T, p)$ and (E-H) $\epsilon_1(T, p)$ contour plots at various frequencies with the same color code. Changes in the conductivity are marginal whereas the permittivity contrast between the percolation peak and the metallic region strongly reduces with higher frequency. The phase boundaries are identical for the different frequencies.
FIG. S4. $\epsilon_1(p)$ of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ at 10 K and various frequencies as indicated. The peak amplitude increases towards lower frequency up to $\epsilon_1 \approx 10^5$, but the position in pressure remains the same. The slow increase with $p$ below 1 kbar is associated with reduction of the Mott gap $\Delta$ while the sharp peak corresponds to phase coexistence around the first-order transition.

pressure tuning. This demonstrates once more the strength of dielectric spectroscopy to study the physics of phase transitions in correlated materials in multidimensional parameter spaces.

While the data presented in Figs. 1 and 4 of the main part were obtained on sample S2 from Argonne Nat. Lab., our findings are reproduced for sample S1 which was synthesized and grown at the University of Stuttgart. We display $T - p$ contour plots of $\sigma_1$ and $\epsilon_1$ at 700 kHz for sample S1 in Fig. S5. Strikingly, the percolation feature appears in the same region in the phase diagram and even the quantitative details such as $T_{\text{max}}$ and the QWL are in good agreement with sample S2 discussed above and in the main text. Due to the limited number of pressures for sample S1, the phase boundaries are not as sharp.

C. Effect of gap reduction on dielectric response

Both our experiments and DMFT-based modeling display huge enhancement of the dielectric function $\epsilon^d_1$, within the phase coexistence region. On the other hand, it is important to establish if the dielectric response in the insulator will possibly also gain partial enhancement due to the reduction of the Mott gap as the transition is approached from the insulating side. To clarify this issue, we calculated $\epsilon^d_1$ for the uniform insulating state and follow it by reducing $U$ until the Mott gap closes and the insulator becomes unstable at $U = U_{c1}$. We find that $\epsilon^d_1 > 0$ for the insulating phase displays only a minor enhancement,
FIG. S5. To check for possible sample-to-sample variations, we measured sample S1 as well (here shown at 700 kHz) and find similar behavior compared to sample S2 shown in Figs. S3, S4 and the main text. The QWL, $T_{max}$ and $T_{FL}$ of sample S2 were inserted for comparison, yielding a good agreement with the data of S1.

remaining positive but essentially constant all the way throughout the coexistence region (Fig. S6). On the other hand, the metallic solution has $\epsilon_1^e < 0$, across the entire uniform metallic phase, all the way up to $U_{c2}$, where the metal becomes unstable at the right side of the coexistence region. Thus, neither uniform phase displays $\epsilon_1^e \gg 1$, which is the hallmark of percolative phase coexistence. In contrast, only our percolative solution displays a significant enhancement within the coexistence region, signaling directly the pronounced effect of spatial inhomogeneities.

FIG. S6. $\epsilon_1^e$ as a function of $U$ at $T = 0.005W$. The red line is for uniform insulating state; the blue one is for uniform metallic state; the purple dashed line shows the effect of percolation, which is found only within the phase coexistence region.
D. Modification of low-temperature transport due to percolation

Detailed low-temperature DC transport properties of $\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3$ around the Mott transition were also discussed recently in Ref. [S4], where a different interpretation was proposed than the one we advocate in this study. Here we show how these data can be reinterpreted in the light of our percolative phase-coexistence picture.

From a very general perspective, materials can be classified as conductors or insulators by examining the temperature dependence of the resistivity. At low-temperature metals generally display Fermi-liquid behavior, and the resistivity assumes the form

$$\rho_m(T) = \rho_o + AT^2,$$

where $\rho_o$ is the (impurity induced) residual resistivity, and the coefficient $A$ describes the strength of electron-electron scattering. In contrast, insulators generally display simple activation:

$$\rho_i(T) \sim \exp\left(\frac{\Delta}{T}\right).$$

(S2)

The approach to the metal-insulator transition can, therefore, be identified by monitoring the behavior of the parameters $A$ and $\Delta$. In strongly correlated metals, one generally finds behavior known as the "Kadowaki-Woods law" $A \sim (m^*)^2$, where $m^*$ is the effective mass of quasiparticles [S7]. If the metal-insulator transition assumes second order character, then one may try to identify the transition by the divergence of $A$ and the vanishing of $\Delta$, as the transition point is approached, which is the procedure adopted in Ref. [S4]. In contrast, if one has a first-order transition, then the behavior should be qualitatively different within and outside the associated phase coexistence region. In particular, within the DMFT scenario, one expects

$$A(U/W) \sim (U/W - (U/W)_{c2})^2,$$

(S3)

to display a divergence only at the boundary of the phase coexistence region. Since one expects $U/W$ to display simple linear dependence on pressure within the transition region, we plot $A^{-1/2}$ as a function of pressure, using the data reported in Ref. [S4]. As we can see on Fig. S7, the quantity does display the expected linear behavior with pressure at $p > 200$ MPa, but shows a pronounced kink and a steep downturn at lower pressures. Interestingly, the $p = 200$ MPa precisely coincides with where one expects the high-pressure boundary of
FIG. S7. Pressure dependence of the Fermi-liquid coefficient $A$ of the dc resistivity. The red dots are obtained from Ref. [S4], and the full line shows that the quantity $A^{-1/2}$ displays linear pressure dependence, as expected from DMFT theory, but only at $P > 200$ MPa; here, $R_{\text{amb}}$ is the resistance at room temperature under ambient pressure. The dashed line shows the correction expected from percolation theory, within the phase-coexistence region (see text).

the phase-coexistence region, based on the location of the critical endpoint of the first-order line, and the general form of the DMFT phase diagram.

This analysis, therefore, indicates that low-temperature transport displays a qualitative change around $p \sim 200$ MPa, with a pronounced enhancement of the coefficient $A$ at lower pressures, which we associate with the onset of percolative transport upon entering the phase coexistence region. To make this point clear, in the following we present general arguments, within percolation theory, to show that the coefficient $A(x)$ must acquire a concentration-dependent correction within the phase coexistence region; it increases due to the reduced metallic filling fraction and displays a divergence (thus $A^{-1/2}$ vanishing) precisely at the percolation threshold $x = x_c$.

To describe percolative transport in presence of phase separation, one should set up a random resistor network [S8], with a binary distribution of resistors having respective temperature-dependent resistivities corresponding to the two coexisting phases, as given by Eqs. (S1,S2). Then, very general considerations have established [S9] that the global resistivity assumes the scaling form

$$\rho(T) = \rho_m(T)\delta x^{-\mu} F(h(T)/\delta \phi), \quad (S4)$$
where \( x \) is the concentration of the metallic phase, \( \delta x = x - x_c \), and the critical exponents \( \mu \) and \( \phi \) are known from percolation theory [S9]. Here, \( h(T) = \rho_i(T)/\rho_m(T) \), and the scaling function \( F(0) = 1 \). Since at low temperature \( h(T) \sim \rho_i(T) \to 0 \), to leading order \( \rho(T) = \rho_m(T)\delta x^{-\mu} \), and the low temperature transport above the percolation threshold \( (x > x_c) \) retains the \( T^2 \) form, but with a coefficient \( A \) acquiring an \( x \)-dependent correction

\[
A(x) = A\delta x^{-\mu}. \tag{S5}
\]

In other words, the quantity \( A(x)^{-1/2} \) (which we plotted in Fig. S7) is multiplied by the factor \( \delta x^{\mu/2} \) and thus vanishes precisely at the percolation threshold. We emphasize that this correction kicks in only as we enter the phase coexistence region. The resulting behavior is shown by the dashed line in Fig. S7, immediately explaining both the "kink" in the pressure dependence of \( A(x)^{-1/2} \), and extrapolated vanishing of this quantity (thus divergence of \( A \)) precisely close to the first-order transition line, around \( p \approx 140 \) MPa, where we expect the percolation threshold, within our phase-coexistence picture.

We should emphasize that the above scaling arguments are completely general and are firmly established within percolation theory [S9], and do not depend at all on the precise form of the phase coexistence region, or other quantitative details of the problem at hand. It established that the Fermi-liquid coefficient \( A \) acquires a singular enhancement near the percolation threshold, a mechanism that may be overlooked in analyzing experimental data ignoring phase coexistence. It also shows that in presence of percolation, \( A \) will increase faster and display a divergence sooner than what one would find for the corresponding pure metallic phase, which as a metastable state should persist all through the entire coexistence region.

---

[S1] Y. Kurosaki, Y. Shimizu, K. Miyagawa, K. Kanoda, and G. Saito, Phys. Rev. Lett. 95, 177001 (2005).

[S2] M. Pinterić, M. Čulo, O. Milat, M. Basletić, B. Korin-Hamzić, E. Tafra, A. Hamzić, T. Ivek, T. Peterseim, K. Miyagawa, K. Kanoda, J. A. Schlueter, M. Dressel, and S. Tomić, Phys. Rev. B 90, 195139 (2014).

[S3] T. Furukawa, K. Miyagawa, H. Taniguchi, R. Kato, and K. Kanoda, Nat. Phys. 11, 221 (2015).
[S4] T. Furukawa, K. Kobashi, Y. Kurosaki, K. Miyagawa, and K. Kanoda, Nat. Commun. 9, 307 (2018).

[S5] M. M. Qazilbash, M. Brehm, B.-G. Chae, P.-C. Ho, G. O. Andreev, B.-J. Kim, S. J. Yun, A. V. Balatsky, M. B. Maple, F. Keilmann, H.-T. Kim, and D. N. Basov, Science 318, 1750 (2007).

[S6] A. Pustogow, A. S. McLeod, Y. Saito, D. N. Basov, and M. Dressel, Sci. Adv. 4, eaau9123 (2018).

[S7] A. C. Jacko, J. O. Fjaerestad, and B. J. Powell, Nat. Phys. 5, 422 (2009).

[S8] S. Kirkpatrick, Rev. Mod. Phys. 45, 574 (1973).

[S9] D. Stauffer and A. Aharony, Introduction to Percolation Theory (CR Press, Boca Raton, FL, 1991).