Topical Review

Electrodynamics of quantum spin liquids

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Received 29 August 2017, revised 29 March 2018
Accepted for publication 6 April 2018
Published 25 April 2018

Abstract
Quantum spin liquids attract great interest due to their exceptional magnetic properties characterized by the absence of long-range order down to low temperatures despite the strong magnetic interaction. Commonly, these compounds are strongly correlated electron systems, and their electrodynamical response is governed by the Mott gap in the excitation spectrum. Here we summarize and discuss the optical properties of several two-dimensional quantum spin liquid candidates. First we consider the inorganic material herbertsmithite ZnCu$_3$(OH)$_6$Cl$_2$ and related compounds, which crystallize in a kagome lattice. Then we turn to the organic compounds $\beta'$-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$, $\kappa$-(BEDT-TTF)$_2$Ag$_2$(CN)$_3$ and $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, where the spins are arranged in an almost perfect triangular lattice, leading to strong frustration. Due to differences in bandwidth, the effective correlation strength varies over a wide range, leading to a rather distinct behavior as far as the electrodynamical properties are concerned. We discuss the spinon contributions to the optical conductivity in comparison to metallic quantum fluctuations in the vicinity of the Mott transition.

Keywords: electrodynamical, quantum spin liquids, magnetic, frustration, spinons

(Some figures may appear in colour only in the online journal)

1. Introduction

The pioneering work on geometrical frustration dates back to the 1920s, when Pauling [1] realized that the hydrogen bonds between H$_2$O molecules in ice can be allocated in multiple ways. Any given oxygen atom in water ice is situated at the vertex of a diamond lattice and has four nearest-neighbor oxygen atoms, each connected via an intermediate proton. According to the ice rule, the lowest energy state has two protons positioned close to the oxygen and two protons positioned farther away, forming a 'two-in two-out' state. Although these considerations dealt with electric dipoles, Anderson [2–4] mapped them to a spin model possessing an extensive degeneracy of states. Two-dimensional (2D) arrangements and many generalizations have been studied subsequently [5–9].

These so-called resonating-valence-bond (RVB) states do not feature any long-range magnetic order or broken lattice symmetries, but are believed to exhibit non-local, topological order [10]. They can be considered as variational ground states constituted of only the shortest possible valence bonds (singlets), with equal weights for all bond configurations. While the formation of a valence bond implies a gap to excite those two spins, long-range valence bonds are more weakly bound and thus a gapless spectrum is possible in spin-$\frac{1}{2}$ systems. It was suggested that such states can be described by gauge theories and these gauge excitations should be visible in the spectrum [11]. In his seminal paper of 1987 Anderson applied the idea of resonating valence bonds to high-temperature superconductivity in cuprates [12–14].

Pure dipolar interaction has been realized in fermionic and bosonic quantum gases, where large magnetic moments provide the dominant interaction [15, 16] in the absence of any other force. In solids, however, exchange and Ruderman–Kittel–Kasuya–Yosida interactions are of superior importance [17, 18], and there are only few examples of mainly dipolar lattices, such as isolated water molecules in nanopores [19, 20].

While in chains, rectangular and cubic lattices the alternating formation of dipoles can be easily obtained, geometrical
frustration becomes an issue in triangular, hexagonal, kagome and hyper-kagome lattices or tetragonal structures, for instance (figure 1). The situation is more intriguing in the case of quantum spin systems when geometrical frustration and quantum fluctuations may prohibit the formation of long-range order even at the lowest temperatures. In these cases liquid-like ground states are expected, and the numerous investigations of quantum spin liquids and spin ice reflect these efforts [11, 21–23].

It took decades before the theoretical concept of quantum spin liquids was actually realized in solids, first in the organic compound $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, which crystallizes in a triangular pattern [24, 25], and later in the kagome lattice of ZnCu$_3$(OH)$_6$Cl$_2$, [26–28]. Recently, three-dimensional pyrochlore materials, such as A$_2$B$_2$O$_7$ with A = Pr or Ho and $B$ = Hf, Ti, Zr, etc, have been discussed as possible quantum spin liquids with frustration and disorder of superior importance [11, 21, 22, 29]. Still there is a lack of direct experimental confirmation, and also the theoretical description of real systems is unsatisfactory; by now the nature of the spin liquid state must be considered as rather unclear. Is the presence of geometrical frustration sufficient to realize a spin liquid? What is the influence of disorder, always present in actual materials? These compounds constitute layered structures, but how important is the coupling to the third dimension? Naturally, quantum spin liquids are mainly considered from the magnetic point of view and the present status is summarized in several recent reviews [11, 23, 30–32]. But how does the electronic degree of freedom is affected by the formation of a quantum spin liquid? Is it caused by the coupling of spin and charge degrees of freedom, or is it due to geometrical frustration and inherent disorder? Here we want to focus on the electrodynamic properties of 2D quantum spin liquids and review the experimental facts observed by now.

2. Herbertsmithite

Among the inorganic spin-liquid candidates, the rhombohedral herbertsmithite ZnCu$_3$(OH)$_6$Cl$_2$ scored highest in popularity. As depicted in figure 2, the copper ions form an almost perfect $S = \frac{1}{2}$ kagome lattice [26], i.e. corner sharing triangles in the plane with strong antiferromagnetic superexchange $J$ of approximately 200 K. No magnetic order is detected all the way down to $T = 50$ mK [27, 28]. Among the transition metal oxides also other candidates such as the vésigniéite BaCu$_3$V$_2$O$_6$(OH)$_2$ [33] and volborthite Cu$_3$V$_2$O$_7$(OH)$_2$·2H$_2$O [34, 35], and also the hyper-kagome Na$_4$I$_3$O$_8$ [36] or PbCuTe$_2$O$_6$ [37] have come under scrutiny. Still the Zn substituted Cu$_3$(OH)$_6$Cl$_2$ remains the prime candidate for a quantum spin liquid. Specific heat and inelastic neutron scattering experiments did not find indications of a spin gap separating $S = 0$ and $S = 1$ down to 0.1 meV, inferring that the spin excitations form a continuum [28, 38–41]. This important issue, however, is far from being settled, neither from the experimental nor from the theoretical side [42–47]. Among other suggestions [10, 48–51], it was proposed that a gapless U(1) spin-liquid state forms with a spinon Fermi surface and with a Dirac-fermion excitation spectrum of the kagome lattice [52, 53].

As far as the charge degrees of freedom are concerned, herbertsmithite exhibits anisotropic electronic properties due to its layered structure. The strong Coulomb repulsion $U \approx 7$ to 8eV [54] makes it a clear-cut insulator with vanishing electronic conduction in all directions up to room temperature. Optical transmission and reflection measurements reveal a charge-transfer band around 3.3 eV, corresponding to 26600 cm$^{-1}$; between 1 and 2 eV pronounced $d$-$d$ transitions have been identified in good agreement with simple LDA calculations, i.e. local density approximation without electronic correlations considered [54, 55].

At lower frequencies pronounced phonon features can be detected [56, 57], as illustrated in figure 3. From the 54 lattice phonons the ten $E_u$ modes are infrared active for light polarized within the $ab$-plane and seven $A_{2u}$ modes for the polarization $E[\parallel]c$. At low temperatures, an anomalous broadening of the low-frequency phonons is observed. Sushkov et al suggested that magnetic fluctuations in the spin-liquid state might couple to the phonons [56]. This is most pronounced in the low-frequency vibration at 115 cm$^{-1}$, which involves the Cl$^-$ and Zn$^{2+}$ ions leading to a deformation of the kagome layer [57]. Raman studies reveal seven phonon modes in the corresponding spectral range [58, 59]. More interestingly, however, is the broad continuum attributed to scattering on magnetic excitations. When the temperature is reduced, the quasi-elastic scattering intensity decreases linearly in contrast to the exponential drop expected for a spin gap [58].

In general, optical experiments are not sensitive to spinon excitations; based on a U(1) gauge theory of the Hubbard model, Lee and collaborators [60] suggested, however, that the spin excitations in a quantum spin liquid may exhibit a Fermi surface and may also contribute to the optical conductivity [47, 61, 62] and the magneto-optical Faraday effect [63]. Hence, investigations of the electrodynamic behavior could

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*Figure 1.* Two-dimensional lattice structures with a high degree of frustration. (a) Triangular lattice, (b) kagome lattice, and (c) hexagonal lattice, where the number of neighbors is reduced from $z = 6$, to 4 and down to 3.
help to establish the spin-liquid ground state and elucidate the low-energy elementary excitations. As seen in figure 3, besides the vibrational features no charge excitations can be observed in the infrared spectral range, due to the large Hubbard $U$. First THz investigations on ZnCu$_3$(OH)$_6$Cl$_2$ were performed by Gedik and collaborators [64]; they report a power-law behavior of the low-frequency optical conductivity $\sigma(\omega) \propto \omega^{1.4}$ in a limited spectral range (figure 4). At 1.3 THz it crosses over to the tail of the rather strong low-frequency phonon at 90 cm$^{-1}$, corresponding to about 2.7 THz. At low-frequencies a power-law dependence on frequency can be identified as $\sigma(\omega) \propto \omega^{1.4}$ (dotted line). Reprinted figure with permission from [64], Copyright 2013 by the American Physical Society.

temperature is raised to 150 K, i.e. well above the regime where spinon excitations are supposed to be dominant. A final conclusion can only be drawn when the observation is confirmed or supporting evidence presented. In particular, experiments at lower frequencies and temperatures are required. The confirmation of spinon contributions to the optical conductivity smoothly extending down to $\omega = 0$ could be considered as a proof of gapless excitations in kagome materials.

3. Organic dimerized Mott compounds

For the molecule-based systems $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ [24, 25], $\kappa$-(BEDT-TTF)$_2$Ag$_2$(CN)$_3$ [65, 66], and $\beta'$-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ [67, 68] the starting point is rather similar: here molecular dimers with $S = \frac{1}{2}$ form a highly frustrated triangular lattice [69–71] as sketched in figure 5(b). Defining the frustration as the ratio of transfer integrals $t'$ and $t$, the compounds are close to perfect frustration as summarized in table 1. At ambient pressure, no indication of Néel order is observed for temperatures as low as 20 mK, despite the considerable antiferromagnetic exchange of $J \approx 220–250$ K [24, 65, 67]. The origin of the spin-liquid phase is unresolved since pure geometrical frustration should not be sufficient to stabilize the quantum spin-liquid state [72–74]. From heat capacity measurements on $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$, gapless spin excitations have been concluded [75] in contrast to thermal transport data [76]. The dispute is not completely resolved. According to Ng and Lee [62], optical studies should provide important information; if spin excitations in $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ exhibit a Fermi surface, they should show up not only in the thermal conductivity but also contribute to the optical conductivity [47, 61, 62]. Recently it was revealed, however, that disorder is intrinsic to $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ and $\kappa$-(BEDT-TTF)$_2$Ag$_2$(CN)$_3$ [66, 77]; this aspect is crucial for understanding the electrodynamic properties of these materials in general.
and can be calculated by tight-binding studies of molecular orbitals or \textit{ab initio} calculations [69, 70, 86]. The intra-dimer transfer integral is given together with the effective Coulomb repulsion $U$ and bandwidth $W$; the power-law exponents are listed for the lowest temperature, according to equation (2): $\sigma(\omega) = \sigma_0 + \omega^{\beta_1} + b\omega^{\beta_2}$. For comparison, the respective data for herbertsmithite are listed, which forms a kagome lattice [30, 65, 67, 78–83].

For different molecular-based quantum spin-liquid compounds on a triangular lattice with antiferromagnetic coupling $J$ the degree of frustration $t'/t$ is given, together with the effective correlations defined as the ratio of inter-dimer Coulomb repulsion $U$ and bandwidth $W$; the power-law exponents are listed for the lowest temperature, according to equation (2): $\sigma(\omega) = \sigma_0 + \omega^{\beta_1} + b\omega^{\beta_2}$. For comparison, the respective data for herbertsmithite are listed, which forms a kagome lattice [30, 65, 67, 78–83].

Table 1. For different molecular-based quantum spin-liquids compounds on a triangular lattice with antiferromagnetic coupling $J$ the degree of frustration $t'/t$ is given, together with the effective correlations defined as the ratio of inter-dimer Coulomb repulsion $U$ and bandwidth $W$; the power-law exponents are listed for the lowest temperature, according to equation (2): $\sigma(\omega) = \sigma_0 + \omega^{\beta_1} + b\omega^{\beta_2}$. For comparison, the respective data for herbertsmithite are listed, which forms a kagome lattice [30, 65, 67, 78–83].

| Compound                  | $J$ (meV) | $t'/t$ | $U/W$ | $\beta_1$ | $\beta_2$ |
|---------------------------|-----------|--------|-------|-----------|-----------|
| $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ | 19        | 0.83   | 1.52  | 0.9       | 1.3       |
| $\kappa$-(BEDT-TTF)$_2$Ag$_2$(CN)$_3$ | 19        | 0.90   | 1.96  | 0.6       | 1.3       |
| $\beta$-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ & 22        | 0.90   | 2.35  | 1.75  | 4.2       |
| ZnCu$_3$(OH)$_6$Cl$_2$ & 17        | 1      | 5–8   | 1.4   | —         |

Nakamura et al [84] report a significant reduction in the low-energy ($\nu < 700$ cm$^{-1}$) Raman-scattering intensity, when the spin-liquid compound $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ is compared to the antiferromagnetic Mott insulator $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl. They assign these to the two-magnon excitation processes. While in the latter compound phonon anomalies indicate the antiferromagnetic order at $T_N = 27$ K, the spin liquid compound $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ exhibits a change in the phonon lines around 90 K and a drastic drop in intensity when the compound crosses over from the thermally activated semiconducting (sometimes called ‘bad metallic’) to Mott-insulating behavior at around 50 K [85], in agreement with comprehensive electrodynamic and transport studies [83].

3.1. Infrared properties

In contrast to the completely insulating herbertsmithite, where the on-site Coulomb repulsion $U$ is large compared to the hopping integral $t$, the electron-electron repulsion for the dimerized charge-transfer salt is significantly lower: $U \approx 0.5$ eV. As illustrated in figure 6, $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ is very close to the Mott insulator-to-metal transition ($U \approx 1.5$ W) [83]; in fact under hydrostatic pressure of only 4 kbar it becomes superconducting at $T_c = 3.6$ K [24, 88]. For $\kappa$-(BEDT-TTF)$_2$Ag$_2$(CN)$_3$ approximately 10 kbar are needed to enter the superconducting state [65]. In figure 7 we display the infrared conductivity for three organic spin-liquid candidates measured at various temperatures using light polarized within the highly conducting plane. The overall shape of $\sigma(\omega)$ is rather similar and dominated by a broad band that peaks around 1700–2300 cm$^{-1}$, which arises from the electronic transitions between the lower and upper Hubbard bands and is therefore a measure of the effective Coulomb repulsion $U$ [89–93]. The values for $U/W$ extracted from figure 7 are listed in table 1 [83].

Besides these electronic excitations, sharp vibrational features are observed below 1500 cm$^{-1}$. In particular for the $\kappa$-phase BEDT-TTF salts the $A_g$ intramolecular vibrations—supposed to be infrared silent—become infrared active by electron–molecular vibrational (emv) coupling to the charge-transfer excitations. They are shifted down in frequency with respect to the corresponding Raman modes [94–98] and are strongly distorted. Numerical investigations of the electronic ground state and the molecular and lattice vibrations reveal the importance of the anion network for...
the vibrational features extending down to the THz range of frequency [66, 77, 99].

In order to elucidate the low-energy excitations within the Mott gap, the lower frames in figure 7 enlarge the optical conductivity in the far-infrared spectral range. The three compounds exhibit a rather different temperature evolution. For $\beta'$-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$, the compound with strongest effective correlations U/W, a clear Mott gap opens around 125 K and continuously grows up to 650 cm$^{-1}$ when the temperature is lowered to 5 K. Also $\kappa$-(BEDT-TTF)$_2$Ag$_2$(CN)$_3$ behaves like a typical insulator, where the in-gap states are depleted upon cooling [100]. The opposite behavior is found in the case of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ (figure 7(f)), which exhibits a temperature dependence characteristic of metals: the low-frequency optical conductivity increases upon lowering the temperature [101–103]. The far-infrared spectral weight of the weakly correlated spin-liquid compound $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ increases significantly for $T < 100$ K and no clear-cut Mott gap is observed in the optical spectra. Such metallic behavior is unexpected considering the absence of a Drude peak related to coherent transport; at $\omega = 0$ all three compounds—including $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$—are electrical insulators as determined from dc transport [83].

3.2. $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

In order to gain more insight into the low-energy dynamics, in figure 8 the conductivity $\sigma(\omega)$ of $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$—as the best studied example of the organic quantum spin-liquid candidates—is displayed in a wide range of frequencies for both in-plane polarizations and different temperatures between $T = 300$ and 10 K. In the kHz and MHz range, hopping conduction is identified as the dominant transport mechanism, accompanied by a broad dielectric relaxation at lower frequencies that bears typical fingerprints of relaxor ferroelectricity [77, 104, 106–108]. For low temperatures, $T < 50$ K, we find an appreciable increase in $\sigma(\omega)$ of the high-frequency dielectric data, which nicely matches the slope observed in the GHz and THz range. The dashed magenta lines in figure 8 simply interpolate the break in our data by

$$\sigma(\omega) = \sigma_0 + a\omega^\beta,$$

with a temperature-dependent constant $\sigma_0$ and prefactor $a$.

The exponent $\beta$ of the power law is approximately 0.4 and increases to almost 1 when the temperature is reduced below $T = 100$ K, as summarized in figure 10(a). It should be noted that the rise in $\sigma(\omega)$ and the corresponding power-law behavior is already observed above 300 kHz in the low-temperature dielectric data (figure 8). Covering such an extremely broad spectral range leads to a high confidence in the power-law exponents [105].

Interestingly, in figure 7 we do not observe a gradual filling or closing of the gap in $\kappa$-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ but an increase in the exponent $\beta$ of the power-law $\sigma(\omega) \propto \omega^\beta$ as the temperature is reduced. Our robust observation [101, 103] is an unambiguous signature that these low-energy excitations...
The extracted values are in accord with the preliminary analysis of the independent experiments on a different \( \kappa-(BEDT-TTF)_2Cu_2(CN)_3 \) single crystal presented in [103].
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ and κ-(BEDT-TTF)$_2$Ag$_2$(CN)$_3$ at the lowest temperature ($\beta_2 \approx 1.4$), it approaches $\beta_2 \approx 4.5$ for $\beta'$-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$.

In a recent theoretical study Dobrosavljević and collaborators [112] concluded that spinons are not well-defined close to the Mott transition. As soon as the electrons become delocalized, the spins have to follow the charge movement, which destroys the coherence of the postulated spinon Fermi surface. Thus spinons do not play a significant role in the vicinity of the Mott transition and within the high-temperature quantum critical regime above the Mott point, in contrast to previous suggestions [60, 113, 114]. In other words, fingerprints of spinon excitations can only be expected in the optical properties of those spin-liquid compounds, which are located deep inside the Mott state, making sure that the charge response is completely absent. For the examples of the three organic crystals discussed here, $\beta'$-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ is the prime candidate to find evidence for spinon contributions to the conductivity, according to table 1 and figure 6. Even then we have to look at rather small frequencies and temperatures well below the antiferromagnetic exchange coupling $J \approx 250$ K [67].

4. Spinon contribution

By performing THz transmission measurements on $\beta'$-EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ single crystals of various thicknesses at temperatures as low as 3 K, it was possible to cover the range down to 3 cm$^{-1}$ with the required accuracy and directly evaluate the optical conductivity without any extrapolation [115]. As seen from figure 12 for both polarizations within the highly conducting plane, an additional absorption process can be identified that adds to the electronic background [115].

Figure 9. Optical conductivity of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ measured at $T = 100, 50$, and 20 K for the two crystallographic directions, upper and lower panels as indicated. The dashed magenta lines correspond to the power-law behavior extending to low-frequencies, the solid red lines indicate the high-frequency power-laws: $\sigma = \sigma_0 + a_0^\beta_1 + b_0^\beta_2$. The data are taken from [77, 105].

Figure 10. (a) Temperature dependence of the power-law exponents $\sigma_1(\omega) \propto \omega^\beta$ of κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ for $E \parallel b$ (solid symbols) and $E \parallel c$ (open symbols). (b) The crossover frequency $\omega_c$ between both regimes shifts with temperature corresponding to $\hbar \omega_c \approx k_B T$. The error bars (shown only for the $c$-polarization) indicate the uncertainty in the power-law fit and the determination of the crossover frequency, respectively.

According to figure 11 the underlying conductivity solely due to charge excitations following a power law $\sigma_1(\omega) \propto \omega^{1.75}$ in the
far-infrared range, crossing over to a steeper slope where the tail of the Mott–Hubbard band dominates. Below approximately 200 cm\(^{-1}\), however, the optical conductivity exceeds the power-law extrapolation considerably, giving evidence for an additional contribution to the electronic response. At much lower frequencies, \(\sigma_1(\omega)\) gradually levels off toward the dielectric data \[116\], comparable to what is plotted in figure 8 for \(\kappa\)-(BEDT-TTF)\(_2\)Cu\(_2\)(CN)\(_3\). The transition regime is consistent with a \(\sigma_1(\omega) \propto \omega^2\) behavior as suggested by Ng and Lee \[62\], that catches up the interpolated value of the correlated electrons rather quickly. Although for \(T \to 0\) this range is enlarged to lower energies, as hopping conduction freezes out, the \(\omega^{1.75}\) decay is approached asymptotically in the GHz range. This gives the lower bound of the spinon-dominated optical conductivity. Thus, the realm of coherent spinons is limited to the range from MHz frequencies to 200 cm\(^{-1}\). When the electronic conductivity exceeds the spinon contribution, the related Fermi surface is eventually damped away, as discussed in \[112\].

Now we can subtract the smooth electronic background determined in figure 12 and focus on the excess conductivity, that is seen in the linear plot of figure 13 as a broad feature...
of the optical conductivity below 200 cm⁻¹. Apart from a few vibrational features, it is rather isotropic and confined to a frequency range determined by the antiferromagnetic exchange $J \approx 250$ K = 175 cm⁻¹. This dome-shaped in-gap absorption is naturally attributed to spinons, which occur when $J$ is the dominant energy scale and the electronic conductivity is sufficiently suppressed at low temperatures. In the static limit, $\sigma_1(\omega)$ decays faster than the $\omega^{1.75}$ power-law background of the optical data. Thus spinons affect neither the optical range nor the dc response where the physics of correlated electrons prevails; nevertheless they can be observed at finite temperatures in a limited frequency range well below the Mott gap.

Going back to the overview on several quantum spin liquids plotted in figure 6, we can now understand why for $\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3$ no indications of spinons could be seen in the optical conductivity (figure 9) [101, 103]. Due to the weaker correlations $U/W$ the compound is located much closer to the insulator-to-metal phase boundary [83] and consequently the tail of the Mott–Hubbard excitations decays much slower towards $\omega \to 0$. $\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3$ exhibits a power-law conductivity with a weaker slope and larger absolute value compared to $\beta'-(\text{EtMe}_3\text{Sb})\text{Pd}(\text{dmit})_2$ [2]. Hence, the electronic contribution to the electrodynamic response of $\kappa-(\text{BEDT-TTF})_2\text{Cu}_2(\text{CN})_3$ dominates well into the GHz range of frequency.

5. Summary

In all cases that came under scrutiny here, the power laws observed in all spin-liquid compounds are not in accordance with the theoretical values predicted for spinon contributions [62], neither at low, nor at high frequencies. In the low-frequency limit, the exponent $\beta_1$ generally resembles Jonscher’s universal power law of the dielectric response [117] as it is widely observed in disordered solids. At high frequencies and temperatures spinons should not play a role because the excitation energy exceeds the antiferromagnetic exchange coupling $J$. The frequency and temperature dependence observed in the optical conductivity of these quantum spin liquid compounds is governed by charge excitations rather than magnetic contributions. For systems with a large Coulomb gap, such as $\beta'-(\text{EtMe}_3\text{Sb})\text{Pd}(\text{dmit})_2$ or herbertsmithite, the spinon contribution becomes detectable in the GHz and THz ranges at very low temperatures when the in-gap absorption is significantly reduced. Here we can identify a contribution to the $\omega \to 0$ optical conductivity that can be assigned to gapless spinon excitations.

Acknowledgments

Over the years, we experienced fruitful collaborations with several groups in the various fields. We acknowledge the useful collaboration with and contributions from V Dobrosavljević, S Fratini, B Gorrshunov, R Hübner, T Ivek, R Kato, C Krellner, Y Li, A Löhle, I Mazin, M Pinterić, P Puphal, R Rösellhuber, G Saito, J A Schlueter, R Valenti, S Tomić, Y Yoshida and E Zhukova. The work was supported by the Deutsche Forschungsgemeinschaft (DFG) via DR228/39-1 and DR228/52-1.

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