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Can one hear the lineshape of a quantum drum?

Observation of a marginal Fermi glass using THz 2D coherent spectroscopy

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Recommended with a Commentary by S.A. Parameswaran, Rudolf Peierls Centre for Theoretical Physics, University of Oxford

The title of this article is inspired by a classic problem in mathematical physics posed by Mark Kac [1] over fifty years ago: *Can One Hear the Shape of a Drum?* Kac was interested in whether one could infer the shape of a fluctuating membrane from its spectrum of normal modes. Spectroscopy in quantum condensed matter poses a somewhat different challenge. Here, one is interested inferring the properties of the low-energy theory from experimentally measured features of the excitation spectrum — such as its lineshape and evolution with parameters. However, in contrast to single atoms, the spectrum of a macroscopic system is typically complicated and difficult to characterize. Disentangling spectroscopic response relies on various simplifying features. For example, in translationally-invariant systems momentum-resolved scattering experiments can separate out the dynamics at distinct wavevectors. In the linear-response regime, this amounts to measuring the dynamical structure factor $S(k, \omega)$ and allows us to infer, for instance, the dispersion $\omega(k)$ of quasiparticles or collective modes. Much theoretical effort has been expended in developing tools to compute such observables in both microscopic models and low-energy effective theories.

However, there are several cases where such simplifications are impossible. An immediate challenge is posed by disordered systems, where translational invariance is absent. Another subtlety emerges in strongly correlated systems, especially those purported to host fractionalized excitations: the probing external field cannot create single fractionalized quasiparticles, but instead creates several at a time (A less exotic example is the ordered phase of the 1D quantum Ising model, where the natural excitations are domain walls in the Ising order parameter, but probing fields can only create spin flips that are bound states of two domain walls.) A third complication is probe-specific: optical response in the THz regime relevant to many solid-state systems is usually restricted to zero wavevector, since the corresponding wavelength typically exceeds the natural length scales of the problem.

To see why the conventional strategies are not particularly helpful in these settings, it is useful to think about how strong quenched disorder and fractionalization would manifest in linear response. Both these phenomena 'broaden' the response, but for very different

reasons. Recall that broadening can be either "inhomogenous" — due to the existence of many distinct sharp modes with a spread in frequency — or "homogeneous", due to the finite lifetime of excitations. While both types of broadening are present in both cases, the mechanisms responsible are distinct. In a fractionalized system (assuming weak disorder) the inhomogenous broadening is primarily due to the quasiparticle dispersion; the homogeneous broadening is due to the decay of a 'bare' excitation created by the probe into multiple quasiparticles. In contrast, in the disordered system inhomogenous broadening is due to the random distributions of couplings, whereas the homogenous broadening is typically due to interaction effects. Linear response shows no particularly sharp spectral features in either case, and typically cannot distinguish inhomogeneous and homogenous broadening. Ideally, one would seek a probe that distinguishes the two types of broadening clearly, in order to reliably identify the mechanisms responsible for each.

The authors of the recommended paper describe an approach that meets some of these challenges, in the specific setting of a three-dimensional doped semiconductor. Inspired by ideas that have long been applied to magnetic resonance [2] and in physical chemistry [3], they use the technique of 'multidimensional coherent nonlinear spectroscopy'. Here, the multiple dimensions in question are in the *time* domain, and the nonlinearity reflects the measurement of multi-point correlations, i.e. those that lie beyond linear response (that measures two-point correlations). The system is addressed by a sequence of pulses, and the response is measured as a function of the time delay between the pulses and between the final pulse and measurement. The resulting multi-time correlator is then analysed in Fourier space. Although it is evident that this can be applied with an arbitrary number of pulses — allowing in principle any number of 'time' directions — the authors consider the simplest nontrivial example of "two-dimensional" coherent spectroscopy (2DCS). This has the notable feature of being able to disentangle homogeneous and inhomogeneous broadening. To see why, it is helpful to go through the exercise of computing the nonlinear 2DCS of a toy two-level system (TLS), that could variously describe the states of a single spin, or empty and occupied states of a localized electron orbital, etc. For specificity, let us take the TLS to have Hamiltonian $H_0 = \frac{\Omega}{2}\sigma^z$ (σ^μ are the Pauli matrices). It is probed by a pair of delta-function pulses A and B at times 0 and τ, and at a time t after the second pulse its state is measured. Each pulse (that we take to be $\propto \sigma^x$) causes transitions between the ground and excited states, effectively rotating the TLS 'spin' on its Bloch sphere by an angle $\theta_{A,B}$ that depends on the pulse strength. The response can be computed by expanding in powers of $\theta_{A,B}$, and it straightforward to verify that only *odd* powers contribute in the case when the measuring field is of the same symmetry as the pulses. The terms proportional to $\theta_{A,B}$ are linear responses to each pulse in isolation. At third order, however, one finds several distinct contributions, of which two are particularly relevant once dissipation is incorporated. The first, "pump-probe response" corresponds to when the B-pulse measures linear response in a state $|\psi_A(\tau)\rangle = e^{-iH_0\tau}\sigma_x|0\rangle$ driven out of equilibrium by the the A-pulse* The second "rephasing" response is reminiscent of a spin echo, in that the phases between pulses A and B exactly cancel when $t = \tau$. Absent dissipation, Fourier transforming the response places these two contributions at distinct positions in the (ω_t, ω_τ)-plane: the pump-probe signal is

*However, in contrast to standard pump-probe experiments in 2DCS the *macroscopic system* is usually not driven into a non-equilibrium state.

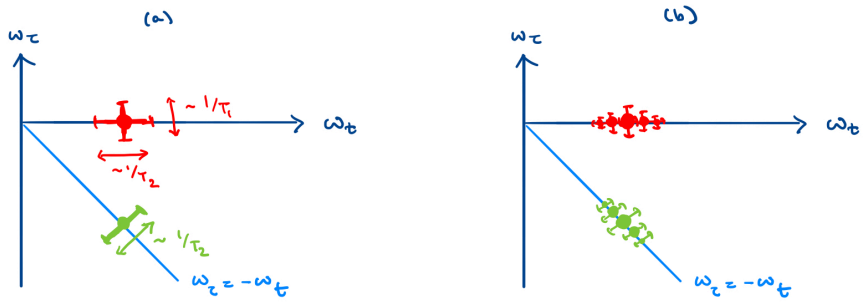

Figure 1: Sketch of Fourier-space 2DCS for (a) single TLS and (b) collection of TLSs.

a δ-peak at $(\pm\Omega, 0)$, whereas the rephasing signal appears at $(\pm\Omega, \mp\Omega)$. Once dissipation is included, the two signals broaden in different ways. The pump-probe signal is affected both by energy relaxation ("T_1 processes", which broaden it along ω_τ), and by phase relaxation ("T_2" or decoherence processes, which broaden it along ω_t), whereas rephasing (as befits an echo response) is only broadened by T_2 processes, in the $\omega_t = \omega_\tau$ direction [Fig. 1a]. (I have elided some subtleties to do with Fourier transforms and the ordering of pulse sequences.)

The utility of the 2DCS response lies both in the distinctive relaxation channels of these signals as well as their placement and evolution, and is exemplified most clearly in a system whose response can be modeled as that of multiple weakly-coupled TLSs with distinct frequencies (as the authors suggest is true of the material studied in the paper). The summed response will consist of a locus of peaks in the (ω_t, ω_τ) plane, with the pump-probe signal evolving along $\omega_\tau = 0$ and the rephasing signal along $\omega_t = -\omega_\tau$. The extent of these loci is a measure of the bandwidth of inhomogenous broadening, while the transverse width of the peaks allows the extraction of T_1 and T_2 times, and (in principle, and with sufficient energy resolution) any nontrivial dependence of these on the TLS splitting. The intensity distribution along the loci is linked to the frequency distribution of the TLSs, and hence to the low-energy density of states accessed by the probe. [This is sketched in Fig. 1b.]

In the paper in question, the authors apply this technique to phosphorous-doped silicon (Si:P). At doping levels such that the system is in an insulating phase, they perform 2DCS spectroscopy using THz optical radiation. Using the technique described above they extract, for the first time, the behaviour of T_1 and T_2 times and their evolution as the system is doped towards the metal insulator transition (MIT). Both relaxation rates are linear in the excitation frequency; this likely rules out a phonon-dominated relaxation mechanism (which would give a relaxation rate $\propto \omega^3$). They also find strikingly counter-intuitive temperature and doping dependences: for instance, T_1 increases with increasing temperature; both relaxation times *increase* as the MIT is approached. These results are counter-intuitive: one would expect energy relaxation rates to increase (and hence relaxation times to decrease) with increasing temperature, and also that relaxation is faster in the metal (in contrast to the trend near the MIT). The temperature dependence is also inconsistent with phonon relaxation.

By analysing various relaxation mechanisms and hopping processes, the authors arrive at an intriguing possible explanation of these results. First, they posit that the 2DCS spectrum can be mapped on to a picture of TLSs — on the face of it, surprising in a long-range Coulomb interacting system. The basic idea involves interactions in an essential way: near the chemical

potential energy μ, interactions suppress vacant and doubly-occupied localized sites in favor of a singly-occupied sites that extend to about an interaction energy about μ. (Aficionados will recognize that the same mechanism is responsible for the famed 'Coulomb gap' [12] for single-electron tunneling.) This results in a sparse network of localized orbitals; some of these orbitals will be close enough to hybridize. The two states of the TLSs correspond to those where an electron is localized on one or the other orbital, so that each TLS corresponds to a localized particle-hole pair. As a result of the Coulomb interactions, such pairs interact in a dipolar fashion with each other. The 'reshaping' of the density-of-states of resonant pairs by interactions is crucial: the authors argue that this gives an roughly uniform density of pairs about the chemical potential (in contrast to a weakly interacting system, where the density of pairs vanishes as μ is approached). A calculation of relaxation within this model then yields a linear-in-ω relaxation rate, in contrast to the ω^2-dependence in the Fermi liquid. Within this picture, the authors explain the temperature-dependence of T_1 as a consequence of thermal suppression of quantum tunneling preceding the onset of thermally activated hopping. Finally, they argue that the doping dependence of relaxation rates is also consistent with a scenario where interactions dominate hopping: since doping moves orbitals closer together, within the resonant-pair model it favors hopping (which is exponentially sensitive to the separation between orbitals) over interactions (which are only algebraically supressed), so that the relaxation rate would *decrease* as the MIT is approached. The authors dub this phenomenology that of a 'marginal Fermi glass': the term is used in analogy with marginal Fermi liquids, but is here taken it to refer to the fact that the lifetime of particle hole pairs diverges too poorly at low frequencies for them to be truly well-defined excitations. This suggests that there is no simple adiabatic continuation to a non-interacting limit.

It is worth injecting a note of caution: while compelling, the calculations that yield this phenomenology are (as the authors note) poorly-controlled near the MIT, where the experiments are performed. However, the paper does make a persuasive case for the need for any theoretical explanation to invoke both disorder and interactions, with its supplement offering a detailed critique of alternative explanations. The results and the material are intrinsically interesting: Si:P is a classic system that spawned Anderson localization, and the implication that it may host an intrinsically interacting electron glass and the possible implications for many-body localization [4] and related phenomena are both compelling in their own right. The behavior at the MIT itself is also likely rich and worthy of further study. However, to me the real excitement in this work lies in the promise of 2DCS as a new spectroscopic probe of quantum many-body systems that provides qualitatively new information inaccessible to standard probes. While it is a textbook technique in chemistry [5, 6], its use in condensed matter is relatively recent [7, 8, 9]. The prospect of applying 2DCS more widely in condensed matter settings raises a new set of challenges and questions. As demonstrated in the present work, describing the 2DCS response even in systems where an approximate TLS description is possible involves three nontrivial ingredients: (i) identifying the low-energy TLSs, (ii) characterizing their distribution in energy and their couplings to external fields; and (iii) understanding relaxation processes. Evidently, in an interacting many-body system these can be related to each other in an intricate fashion that can significantly alter the 2DCS portrait. Furthermore, low-temperature relaxation in strongly-correlated systems remains poorly understood in many cases. The situation is even more complicated in cases where such an effective TLS picture is not applicable. Very recent theoretical calculations [10, 11]

have explored some special examples, but there is much scope for further work.

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