systems have classical ordered phases at low temperature. Now, Banerjee and co-workers report an interesting development in the search for the ideal Kitaev compound, in looking at the α-$RuCl_3$ system[10]. Using inelastic neutron scattering, they find a number of excitations with a temperature and frequency dependence consistent with a large Kitaev interaction. α-$RuCl_3$ also orders at low temperature, but its Heisenberg term is estimated to be smaller than in the iridates, making it perhaps the best example found so far of a system with a dominating Kitaev interaction. At low energies, the spectra appear to reflect the broken symmetry state that is expected for an ordered state proximate to the Kitaev phase. At higher energies, the spectra more closely resemble that predicted for the Kitaev spin liquid. This partitioning of energy scales is similar to that found in magnetically ordered quasi-1D spin-chain systems such as $KCuF_3$, where the high-energy behaviour reflects that of the free fractionalized spin-1/2 particles of the spin liquid state inherent to strictly 1D, but at low energies there is a crossover to spectra that reflect the ordered state[11].

Going forward, it is important to find new materials or configurations that may reflect even closer idealities of exactly solvable models such as the Kitaev model, or systems that may surprise us in being realizations of topological QSLs. It is also imperative for the community to find new ways of probing these systems that reveal their underlying non-trivial nature in a more explicit fashion. ❐

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GIANT RYDBERG EXCITONS

Probing quantum chaos

Giant Rydberg excitons reveal signatures of quantum chaotic behaviour in the presence of time-reversal symmetry breaking enforced by the background solid-state lattice, and they provide a new mesoscopic platform for fundamental studies of quantum chaos.

Elena A. Ostrovskaya and Franco Nori

Quantum chaos refers to the behaviour of quantum systems whose classical counterparts exhibit random behaviour due to extreme sensitivity to their initial conditions[1]. Classical chaos is most evident in the properties of the system's trajectories, whereas quantum chaos manifests itself in the energy spectra. A regular quantum system exhibits a Poissonian distribution of nearest-neighbour spacings between energy levels, meaning that the energy eigenvalues behave like a sequence of completely independent random variables[2]. In contrast, the eigenvalue statistics of a quantum chaotic system is well described by a suitable Gaussian ensemble of Hermitian random matrices and the system demonstrates strong correlation (repulsion) between its energy levels[3]. Despite the well-developed theory of quantum chaos, experimental observations of its signatures in truly quantum systems are few and far between. Using a giant Rydberg exciton in an exceptionally clean and controllable solid-state environment, Marc Aßmann and colleagues[4] demonstrate the transition from the Poisson distribution of nearest-neighbour energy-level spacings to a distribution governed by the Gaussian unitary ensemble (GUE) statistics, which is a signature of a quantum chaotic system that also has broken time-reversal and all other anti-unitary symmetries.

Since clean, controllable quantum systems that can be used for fundamental studies of quantum chaos are rare, classical wave systems with discrete spectra (such as optical and microwave cavities) have been filling the niche for the past few decades[5,6]. Genuinely quantized systems, such as electrons in quantum dots[7], cold atoms in

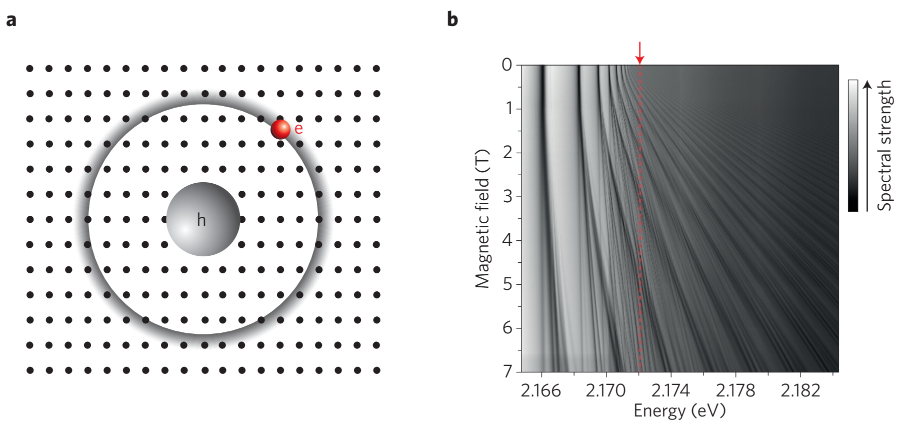

Figure 1 | Complex level structure of a giant Rydberg exciton in a magnetic field. **a**, Schematics of a giant Rydberg exciton where h represents the hole with a large effective mass and e represents an electron. **b**, Transmission spectrum of Cu_2O measured in magnetic fields up to 7 T, starting from the state with principal quantum number $n = 4$. As the magnetic field increases, so do the energy of states and the level mixing, triggering the transition to quantum chaos, as confirmed by the GUE level spacing distribution extracted for the energies above the vertical red dashed line. Figure adapted from ref. 4, Nature Publishing Group.

optical traps[8] and exciton–polaritons in semiconductor microcavities[9] have also been investigated, but exclusively in the context of 'billiards' — resonators for matter waves, which do not always provide access to sufficiently large energy-level statistics. The latter point is important because quantum chaos cannot be deduced from the local behaviour of the energy levels; for example, the local repulsion of two energy levels, that is, avoided crossings, is not a signature of chaos by itself.

The study by Aßmann and co-workers[4] puts forward a new candidate for in-depth studies of quantum chaos: the weakly bound electron–hole pairs in a naturally occurring copper oxide Cu_2O crystal, termed giant Rydberg excitons[10], which extend over more than tens of billions of solid-state lattice sites (Fig. 1a). The extent and composition of this mesoscopic quantum system is analogous to a highly excited hydrogen atom, whereby the hole (having a large effective mass in the crystal) plays the role of a proton. Indeed, the Bohr radius of these giant excitons (~2 μm) is comparable to that of Rydberg atoms with a high principal atom number, the latter being well suited for studies of quantum chaos[11] since the relatively slow, nanosecond-scale orbital periods allow for electromagnetic manipulation and probing of electron states.

This commonly used analogy cannot be extended too far. The main difference between the Rydberg atoms and excitons stems from the background solid-state lattice and, in particular, from interactions of excitons with phonons (lattice excitations). For example, the crystal environment leads to a small Rydberg energy (90 meV) that allows one to reach the quantum chaos regime at magnetic field strengths that are orders of magnitude smaller than for atoms[4] (Fig. 1b). Most importantly, the phonon-mediated scattering of optically excited excitons obeys strict momentum selection rules and leads to a generalized time-reversal symmetry breaking. This, in turn, strongly modifies the statistics of energy eigenstates in the chaotic regime, and causes a transition from the chaotic energy-spacing distribution, governed by the Gaussian orthogonal ensemble (GOE), to that governed by the GUE (ref. 12). The difference in physical properties of the system's Hamiltonian is significant because of the symmetry breaking, but its spectral manifestation is very subtle: the level repulsion (probability of finding energy levels separated by increasingly small intervals) is quadratic for the GUE and linear for the GOE. Although for most physical systems it is impossible to differentiate between the two regimes in an experiment, the exceptionally clean environment of the Cu_2O crystal allowed Aßmann and his colleagues to do so (see Fig. 2).

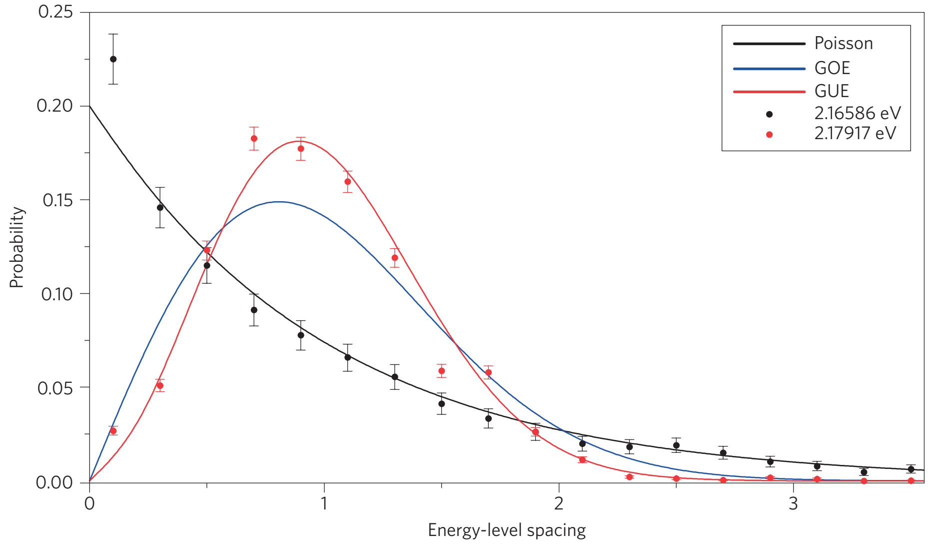

Figure 2 | Nearest-neighbour level spacing distribution in the regular and chaotic regions, compared with the predictions for the Poissonian ensemble (black), GOE (blue) and GUE (red curve). The GUE distribution clearly provides the best fit to the experimental data in the chaotic regime. For small energy-level spacings, the GOE and GUE distributions are linear and quadratic, respectively. A Poissonian counting error defines the error bars. Figure adapted from ref. 4, Nature Publishing Group.

The unambiguous delineation between the GOE and GUE statistics achieved for the giant Rydberg excitons is due to the fact that their transmission spectra can be probed with unparalleled precision, and around 3×10^6 energy states can be optically resolved with spectrally filtered white light. This results in massive spectral statistics that are not feasible, for example, for microwave or optical billiards. Nevertheless, the obvious shortcoming of the measurement shown in Fig. 2 is that a perfect fit to the theoretical GUE distribution is expected only for a constant mean density of states (DOS), which is not the case in the experiment. To remedy this, Aßmann and colleagues use the experimental data to calculate the mean value of the 'two-gap' correlation function, which is a measure of consecutive spacings between three neighbouring levels, and is less sensitive to variations in the DOS. This alternative measure confirms the GUE statistics of energy levels[4].

The opportunities offered by giant Rydberg excitons in moderate magnetic fields extend well beyond the fundamental studies of the transition to quantum chaos in a well-controlled parameter space. Indeed, the very same interaction with the crystalline environment that ensures non-trivial breaking of time-reversal symmetries could allow non-Hermitian spectral degeneracies[6,9] (exceptional points) to be explored at low field values. Furthermore, the spin–orbit interaction, which leads to the fine structure of energy levels in magnetic fields, should be significant for this system, and could lead to non-trivial topological properties of the eigenstates. These exciting effects could be revealed by further spin-resolved experiments. ❐

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