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Inducing new material properties with HYBRID LIGHT–MATTER STATES

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Even in the absence of light, coupling cavities with molecules and materials can modify their chemical reactivity, conductivity, and more.

The link between light, matter, and vacuum has been a subject of discussion since antiquity. The conceptual revolution that started in the 1920s with quantum mechanics and led to the development of quantum electrodynamics (QED) changed our understanding of light–matter interactions—particularly the role of vacuum, the space through which light propagates.

In the 1970s a series of milestone experiments by atomic physicists showed that optical cavities could fundamentally modify the spontaneous emission of photons from excited atoms by either enhancing or suppressing it. The effect had already been predicted by Edward Purcell in 1947 and is best understood by considering that a cavity modifies the density of optical states of vacuum. Atoms can emit light only into available optical states. In a cavity the density of those states is altered, and therefore so is the probability of photon emission. Such a system can even enter a regime in which a spontaneously emitted photon is periodically reabsorbed by the atom itself; the resonant frequency of that exchange is known as the Rabi frequency. (For more on cavity QED, see the article by Serge Haroche and Daniel Kleppner, PHYSICS TODAY, January 1989, page 24.)

Such periodic energy exchange naturally results in the formation of hybrid states known as polaritonic states. Their nature, a kind of half-photonic and half-matter chimera, raises the prospect of changing material properties. The most striking aspect of the so-called strong-coupling regime is that the existence of the polaritonic states does not require the cavity to be initially populated with photons. In other words, the coupling can occur with an electronic, vibrational, or other material transition and proceeds via the zero-point fluctuations of the electromagnetic energy inside the cavity.

The optical excitation of such a coupled system by a light source results in the formation of a polariton—a quasiparticle formed when a photon is coupled to a matter excitation. Exciton polaritons have attracted a lot of interest for their ability to, among other things, form condensates that exhibit superfluidity and host vortices (see the article by David Snoke and Peter Littlewood, PHYSICS TODAY, August 2010, page 42).

Here, in contrast, we consider the situation in which coupled systems are not optically excited. Numerous studies over

A microfluidic Fabry–Perot optical cavity for doing quantum electrodynamic chemistry. (Photo courtesy of Thomas W. Ebbesen.)

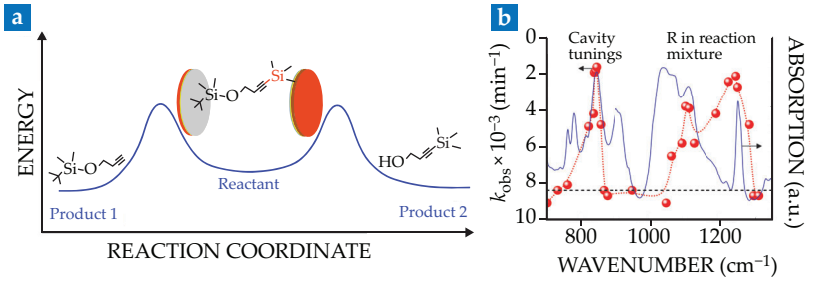


FIGURE 1. CHEMICAL REACTION biasing. **(a)** When the reactant *tert*-butyldimethyl{[4-(trimethylsilyl)but-3-yn-1-yl]oxy}silane is attacked by a fluoride ion, one of two products emerges. If the reaction proceeds in a cavity, it can be biased toward the normally unfavored product by tuning the cavity's resonance to match the vibrational frequency of one of the molecule's bonds. **(b)** The overall reaction rate k_{obs} (red dots) dips when the cavity is coupled to the reactant's silicon–carbon bond (842 cm^{-1}), silicon–oxygen bond (1110 cm^{-1}), or silicon–methyl bond (1250 cm^{-1}). The reactant's IR absorption spectrum (blue line) has the corresponding maxima. (Adapted from ref. 4.)

the past decade have shown that, surprisingly, material properties can be significantly modified under such conditions. In the following sections, we give a few examples of recent findings in chemistry and condensed-matter physics that illustrate the potential of strong coupling in the absence of light to control the properties of matter.

Cavity chemistry

The demonstration that strong coupling to an electronic transition can modify a photochemical reaction has stimulated many theoretical studies.[1] They have shown, for instance, that the experimental observations could be explained by the formation of a polaritonic potential-energy surface (PES) that modifies the internal dynamics and the reactivity landscape.[2]

Polaritonic states are not limited to electronic transitions; they can also be made by coupling a cavity mode with a molecular vibrational transition. Such vibrational strong coupling (VSC) yields a new type of state, the so-called vibropolaritonic state, which has remarkably strong effects on chemical reactivity. Molecular vibrational transitions have relatively high frequencies ω_v, which are fixed by the bond strength f (typically on the order of 10^3 N/m) and by the tiny atomic masses involved in the vibrations. Their resonances are characterized by small Boltzmann factors, $n_v \sim e^{-\hbar\omega_v/k_BT} \sim 10^{-4}$, which describe the probability of being in an excited state. Such molecular modes are therefore in their ground states, even at room temperature.[3]

Chemistry experiments under VSC can be run inside microfluidic optical cavities (see opening image) that are tuned to the IR vibrational bands of the solute, the solvent, or both. A simple Fabry–Perot cavity comprises two mirrors that are separated by a thin polymer spacer, typically about 10 μm thick. The cavity's frequency ω_c can be fine-tuned to a given vibrational transition simply by squeezing the spacer with a screwdriver and monitoring the spectrum with a standard Fourier-transform IR spectrophotometer. The two reactions described below illustrate the consequences of VSC on chemistry.

Most chemical reactions lead to several products because of the complexity of the PES of the reactivity landscape. For practical purposes—lowering cost, reducing waste, and minimizing purification steps—much effort is put into finding ways to selectively orient a reaction toward the desired product, and thus achieve the highest yields. The requisite biasing has traditionally been done by chemical means, such as through the choice of reaction pathway, catalyst, and solvent.

A study of a simple reaction with two possible outcomes, shown schematically in figure 1a, demonstrated that VSC could produce similar biasing toward a particular product. In the absence of VSC, the proportion of product 1 to product 2 was 60:40; under VSC, it became 20:80. That shift reflects modifications in the relative barrier heights.[4] The inversion of the selectivity occurred only when three of the numerous vibrational modes were coupled by tuning the cavity across the various vibrations (see figure 1b). The vibrations corresponded to the silicon–carbon, silicon–oxygen, and the silicon–methyl stretching modes that are associated, not surprisingly, with the bonds being broken in the reaction. Tunable VSC can thus provide information on the mechanism of the reaction.

As can also be seen in figure 1b, VSC slowed down the reaction rates for both products by changing the energy-barrier heights leading to each one. Temperature studies measuring the enthalpy and entropy of activation showed surprisingly large changes in those values, typically an order of magnitude greater than energy associated with the resonant frequency shifts caused by VSC, which are on the order of k_BT. In other

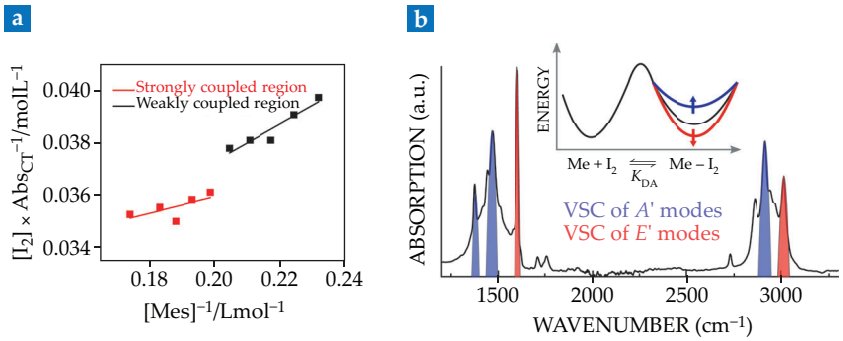


FIGURE 2. SYMMETRY and vibrational strong coupling (VSC). **(a)** The highly symmetric molecule mesitylene can react with iodine through a charge-transfer complexation reaction. A so-called Benesi–Hildebrand plot reflecting the change in the inverse mesitylene concentration versus the charge-transfer complex's absorption reveals an abrupt shift when the coupling transitions from weak to strong. **(b)** An IR absorption spectrum of mesitylene shows its vibrational modes. They can be grouped by the symmetry classes they belong to, *E*' (red) and *A*' (blue). Coupling a cavity to those modes shifts the equilibrium landscape of the mesitylene–I_2 complexation process (inset). Under VSC of the *E*' modes, the equilibrium constant K_{DA} increases and the complex is favored; VSC of the *A*' modes favors the reactants. (Adapted from ref. 6.)

Theoretical concepts of light–matter strong coupling

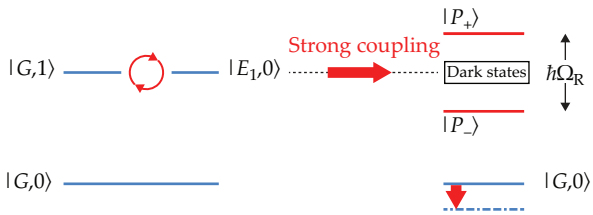
Confining a material in an optical cavity can produce new hybrid light–matter states. When the rate of energy exchange between the material and the cavity—the Rabi frequency—is faster than any dissipative process, the system reaches what's known as the strong-coupling regime. Although strong coupling is difficult to achieve with single entities such as atoms, increasing the number of entities N that are collectively coupled to an optical mode facilitates the process by enhancing the energy exchange.

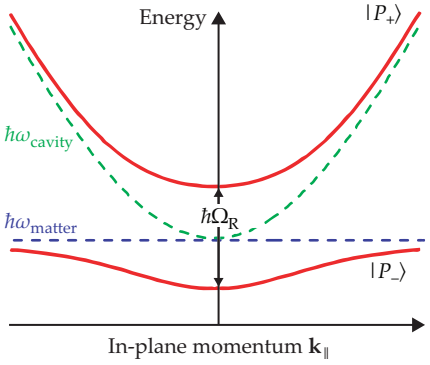
The energetic structure of a strongly coupled light–matter system can be derived simply by describing the matter part as a collective dipole moment, $\mathbf{P} = \sum \mathbf{p}_i$, where $\mathbf{p}_i = \langle e_i|\mathbf{d}|g_i\rangle$ is the transition dipole of one emitter between ground $|g_i\rangle$ and excited $|e_i\rangle$ states. The Hamiltonian describing the coupling between the collective dipole $\mathbf{P}$ and the electric displacement $\mathbf{D}$ of the mode[2] becomes

$$H = H_{\text{cav}} + H_{\text{matter}} - \frac{1}{\varepsilon_0}\,\mathbf{D}\cdot\mathbf{P}$$

In the above framework, the collective ground state for the coupled system is defined as $|G,0\rangle = |g_1, \ldots, g_N\rangle|0\rangle$ with all the emitters in their ground states and no photon, $|0\rangle$, in the cavity. The polaritonic states, $P_\pm = (|G,1\rangle \pm |E_1,0\rangle)/\sqrt{2}$, are formed from the symmetric and antisymmetric superposition of the matter's ground state with one photon, $|G,1\rangle$, and the matter's first excited state with no photon, $|E_1,0\rangle$. They are separated by the enhanced Rabi splitting energy, $\hbar\Omega_R = 2\hbar\Omega\sqrt{N}$, where $\hbar\Omega$ is the interaction strength of a single entity with the vacuum field of the cavity, as illustrated at the top of the figure.

The symmetry of the Hamiltonian





implies that when N oscillators couple to one optical mode, the Hamiltonian has $N+1$ solutions. Two of them, P_+ and P_-, couple to the light field; that leaves $N-1$ states, the so-called dark states that don't couple to the light field but are nevertheless collective and can influence the properties of the coupled system. They should not be confused with uncoupled entities in the cavity, such as those whose dipole moments are oriented perpendicular to the cavity field.

The dispersive behavior of a Fabry–Perot cavity has consequences for the properties of cavity-coupled materials at room temperature. Experiments have shown that it is important to induce strong coupling at the bottom of the dispersion band—the energy versus in-plane momentum plot at the bottom of the figure—to observe changes in material properties.

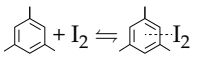
In the case of electronic strong coupling involving molecules, the role of the rotational–vibrational reservoirs associated with each electronic level is crucial and radically differentiates the dynamics of inorganic systems from organic ones. Organic molecules' relaxations are essentially driven by the rotational–vibrational reservoir in the thermal energy scale (k_BT), which implies that as soon as $\hbar\Omega_R > k_BT$, the relaxations of the coupled system cannot be estimated from those of the uncoupled molecular states. Those relaxations occur in a so-called non-Markovian regime, which contributes to some surprising features of the polaritonic states, such as their unexpectedly long lifetimes. (For more on non-Markovian polariton dynamics, see A. Canaguier-Durand et al., *Eur. Phys. J. D* **69**, 24, 2015.)

words, the simple modification of the vibrational frequency spectrum induced by VSC cannot fully account for the experimental observations.

Large activation-energy changes have been systematically seen in all reactions studied to date, regardless of whether VSC decelerates or catalyzes the reaction. Some other fundamental aspect must therefore be involved.[5] The most likely candidate is symmetry, which has been known to play a key role in chemical reactivity since the seminal work of Kenichi Fukui, Robert Woodward, Roald Hoffmann, and Richard Bader in the mid 20th century. (Fukui and Hoffmann shared the Nobel Prize in Chemistry for their work on chemical reactivity; see PHYSICS TODAY, December 1981, page 20.) Symmetry correlations between reactants and products determine the PES of the reactivity landscape and thus affect the products and rates of reactions. The symmetry of the vibrations can also help favor certain pathways over others.

Tilting the balance

A simple charge-transfer complexation reaction between the highly symmetrical mesitylene molecule and iodide illustrates the interplay between VSC and symmetry:[6]



The charge-transfer complex has a distinct absorption band in the UV that can be monitored to extract the kinetic equilibrium constant K_{DA} of the system. The dramatic modifications caused by strong coupling are surprisingly clear: Figure 2a shows an abrupt change in slope, which is reminiscent of a phase transition and reflects changes in both the complex's absorption and the system's K_{DA} when the coupling goes from weak to strong.

When the cavity is tuned across the vibrational bands of mesitylene to induce VSC, the ground-state landscape tilts either toward or away from the product. Notably, the direction of the tilt depends on the symmetry class of the vibration to which the cavity is coupled, as illustrated in figure 2b. The type of vibration, its frequency, and the energy-spectrum shift caused by coupling appear to have little or no impact on the reactivity landscape.

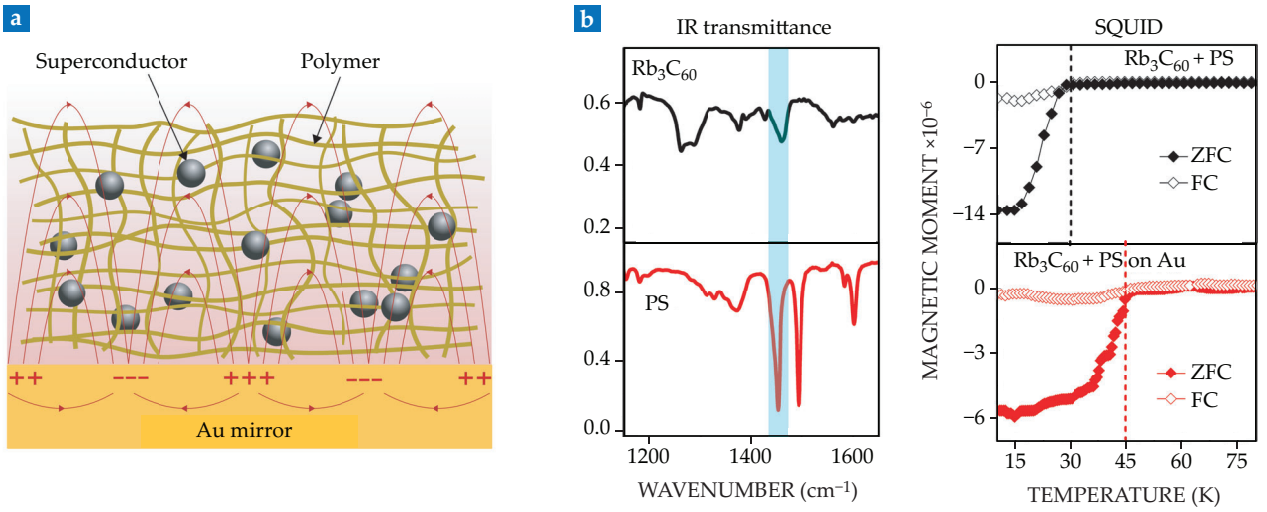


FIGURE 3. SUPERCONDUCTIVITY enhanced by plasmons. **(a)** Coupling the IR absorption band of Rb_3C_{60} nanoparticles to the surface-plasmon modes of a gold mirror can enhance the compound's superconducting properties. A polymer matrix is needed to mediate the coupling. **(b)** Although Rb_3C_{60} absorbs only weakly in the IR, polystyrene (PS) has an overlapping vibrational band that absorbs strongly at the same wavelength and facilitates cooperative coupling. Measurements of the polymer–particle complex's magnetic moment under zero-field-cooled (ZFC) and field-cooled (FC) conditions reveal an increase in the superconductor's critical temperature with surface-plasmon coupling from 30 K to 45 K, and the gaps between the FC and ZFC curves indicate the Meissner effect. (Adapted from ref. 11.)

Although reactivity landscapes can be complex and involve many factors, such as the solvent's properties and steric hindrance, knowing that symmetry affects chemical reactions under VSC should help researchers predict the outcomes of chemical reactions. VSC could even be used to study larger molecules whose symmetries are not as well defined.

Chemistry under VSC, also known as QED chemistry and polaritonic chemistry, is a new approach to the field and simple to implement; it can be done on any chemistry bench in most labs with just a few tools (see the opening image). It is even relatively simple to massively parallelize the microfluidic optical cavities for industrial purposes. The VSC approach is not limited to chemistry. Enzyme activity can also be modified by simply coupling the stretching modes of H_2O, which would open the door to applications to biological systems.[7] Furthermore, since more than one type of molecule can couple to a given cavity mode, it is possible to, for example, quantum mechanically entangle two molecules to the same mode and modify processes such as energy transfer between molecules.[8]

High concentrations of molecules are necessary to achieve strong coupling with cavity modes. Experiments have therefore been limited because chemical reactions are, by contrast, usually carried out in relatively dilute solutions. One way to overcome that problem is by involving the abundant solvent molecules. If the solute and solvent have an overlapping vibrational mode, the cavity can strongly couple to it. For example, putting *para*-nitrophenyl acetate and a solvent that shares its C–O stretching mode together in a Fabry–Perot cavity increased the rate of a solvolysis reaction by an order of magnitude, even though only the solvent was present at a sufficiently high concentration for strong coupling.[9] Theory shows that intermolecular vibrational interactions can indeed mediate such cooperative effects.[10] They are also critical to enhancing the electron–phonon scattering that affects the quantum properties of certain materials, such as superconductors, under strong coupling.

Superconductivity

The shift in molecular vibrational frequencies induced by strong coupling leads naturally to the question of whether phonon-driven solid-state phenomena, such as superconductivity, can be modified in the VSC regime. The challenge of exploring that question lies in the fact that superconductors' IR absorption bands are typically weak and therefore difficult to strongly couple.

The cooperative effect discussed above for solutes and solvents has been used to overcome that limitation. One such experiment tested the well-known superconductor Rb_3C_{60}, whose Cooper pairing is driven by phonons, under VSC.[11] A powder made up of superconductor particles, approximately 200 nm in size, was dispersed in various polymers with different vibrational bands. Spin coating those solutions on gold films (see figure 3a) strongly coupled the samples to the surface plasmons of the metal film. A SQUID (superconducting quantum interference device) magnetometer measured both the critical temperature T_c of each sample and the Meissner effect, a signature of superconductivity.

Of the several polymers tested, only polystyrene had strong vibrational bands that were resonant with the superconductor (see figure 3b). That sample reached the cooperative VSC regime and displayed not only a distinct increase in T_c, from 30 K to 45 K (see figure 3b), but also a clear Meissner effect. In view of the poor signal-to-noise ratio, many samples were tested to confirm the shift in T_c. The increase can be attributed to an enhancement of the electron–phonon coupling discussed above. Further experiments that fully characterize Rb_3C_{60}'s superconducting properties and studies of other superconductors are needed to provide further insight into the consequences of strong coupling on superconductivity.

Electronic transport

When the Rabi frequency of a light–matter system is near the

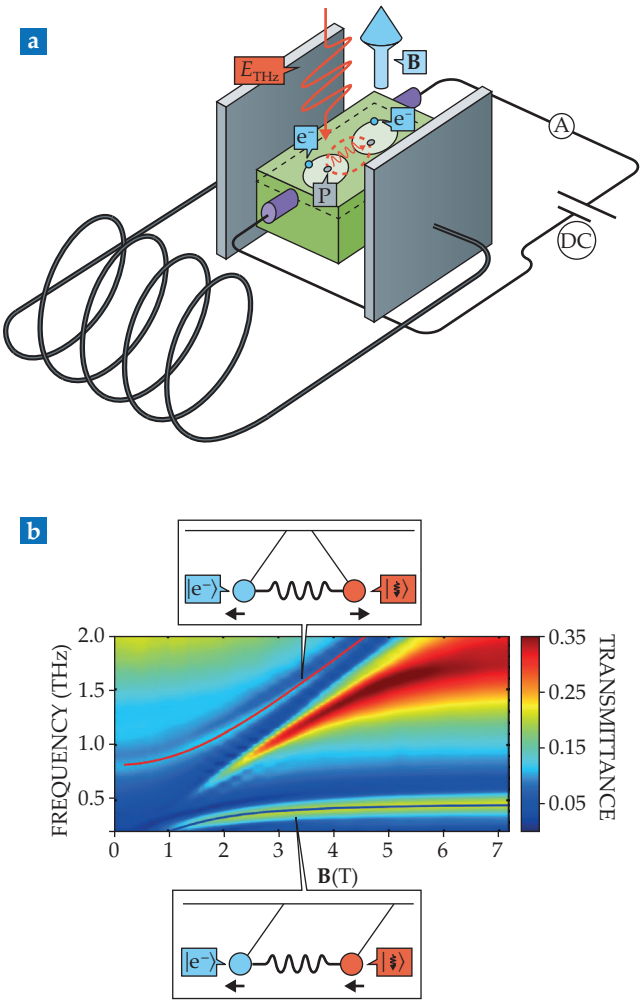


FIGURE 4. STRONG COUPLING in an experimental platform. **(a)** When electrons are trapped inside a heterojunction (green) and subjected to a perpendicular magnetic field **B**, they perform cyclotron orbits. Those orbits can then couple to the vacuum-electric-field fluctuations inside an electronic resonator (here an inductance–capacitance resonant circuit) of deep-subwavelength dimension to create collective excitations of the electron–cavity system known as polaritons. The polaritons can be probed either optically (as shown schematically by the wavy red arrow labeled E_{THz}) or by electrical transport (shown by the ammeter). **(b)** A polaritonic state can be understood as a coupled electron–photon mode whose resonant frequency, shown here in a color plot of the terahertz transmission, is tuned by a magnetic field. Similar to coupled classical pendula, the states exhibit in-phase and out-of-phase resonances. (Adapted from C. Maissen et al., *Phys. Rev. B* **90**, 205309, 2014.)

resonant frequency of the empty cavity, the system enters the ultrastrong-coupling (USC) regime. It can then exhibit some striking features: Its ground state, for example, becomes modified and contains virtual photons.[12] The transition from strong to ultrastrong coupling is naturally gradual, as is the modification of the ground state. We want to probe those changes using electronic transport.

Ultrastrong light–matter coupling can be achieved by engineering both the electronic excitations and the cavity. As already discussed in the context of the chemical reactions, coupling a collective excitation of many dipoles, rather than that of a single atom, to an optical cavity can help the system reach the strong-coupling regime (see the box on page 45). So can a properly designed resonator—for instance, a cavity where the electric field is confined in a deep-subwavelength-sized volume. That level of confinement can be achieved by replacing a Fabry–Perot cavity with an electronic inductance–capacitance resonant circuit.

Using a cavity of deep-subwavelength size greatly enhances the strength of the vacuum-electric-field fluctuations because the same zero-point energy of the electric field, half a quantum, must be squeezed in a smaller volume. Such cavities are most easily fabricated in the terahertz-frequency region, where metals have a very large and negative value of their dielectric function that enables tight confinement of an electric field.

The exploration of the USC regime was predicted to be achievable in two-dimensional electron systems coupled to terahertz cavities,[12] and that prediction was borne out experimentally.[13] In particular, a ratio of order unity between the Rabi frequency of a light–matter system and the resonant frequency of the empty cavity was observed in experiments in which researchers exploited the cyclotron resonance of a 2D electron gas as the matter excitation.[14,15]

Figure 4a illustrates the principle of such a system's operation: A high-mobility 2D electron gas confined by a semiconductor heterostructure is subjected to a strong perpendicular magnetic field **B**. The resulting Lorentz force bends the electron trajectories into quantized cyclotron orbits, and the system displays an equidistant ladder of states, so-called Landau levels, whose spacing is proportional to the field strength B.

Optical transitions between the Landau levels occur in the terahertz-frequency range and can be coupled to an inductive–capacitive circuit of deep-subwavelength dimension; the circuit's capacitor serves as the cavity. The polaritonic states—cavity photons strongly coupled to the cooperative motion of the cyclotrons—appear as resonances that tune with the magnetic field as the Landau level spacing is brought in and out of resonance with the cavity. When the system is probed by an external terahertz excitation, one resonance appears at a frequency higher than that of the bare resonator and another appears at a lower frequency, similar to what happens with a pair of coupled classical oscillators (see figure 4b).

The system illustrated in figure 4 can be used to investigate how light–matter coupling might affect electronic transport. Magneto-transport in a 2D electron system with very low disorder exhibits a rich phenomenology that includes, most famously, the integer and fractional quantum Hall effects. (For more on integer quantum Hall effects, see the article by Joseph Avron, Daniel Osadchy, and Ruedi Seiler, PHYSICS TODAY, August 2003, page 38; on fractional quantum Hall effects, see PHYSICS TODAY, July 1983, page 19.) That richness, however, reflects the theoretical complexity of combining electronic transport and quantum optics, which can make such systems challenging to study.

Initial experiments performed under a weak terahertz illumination that slightly increased the occupation of the polaritonic state showed the role such a state played in the electron-

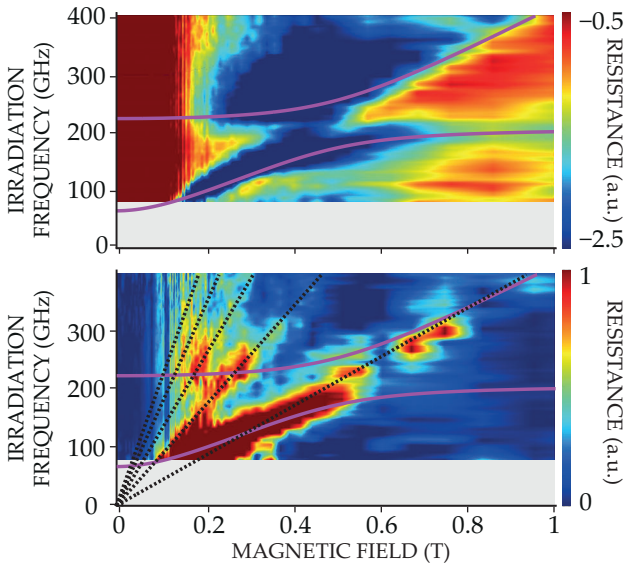


FIGURE 5. MAGNETO-TRANSPORT under weak terahertz illumination. The color plots reveal the change in resistance of a two-dimensional electron gas when probed in the manner illustrated schematically in figure 4. The top graph shows the resistance change for magnetic fields that cause transport to occur in extended states that form polaritons (purple lines). In the lower graph, the Fermi level lies in the localized states that do not participate in the polaritons; the resistance maxima therefore follow the dispersion of the Landau levels (black dashed lines). (Figure adapted from ref. 15.)

transport properties of the 2D electron gas (see figure 5). In the semiclassical limit—at which the presence of the Landau levels has not yet produced the quantized resistance steps characteristic of the integer quantum Hall effect—electron transport was found to be sensitive to the presence of the polaritons when the Fermi level coincided with a Landau level and transport was therefore dominated by extended states. In contrast, when the Fermi level lay between Landau levels in the localized states responsible for the quantum Hall conductance, transport was immune to the presence of the polaritons. The small extent of the states' wavefunctions caused the states to couple only weakly to the polaritons.

The experiments discussed above were conducted at millikelvin temperatures in ultrapure semiconductor heterostructures to minimize the impact of disorder and thermal excitation on the electrons' motions. However, the effects of polaritonic states on transport have also been studied in a completely different regime.[16] In room-temperature organic semiconductors, electronic transport is typically dominated by short-range site-to-site hopping. Experiments have shown that strong coupling can produce a striking increase in charge mobility in the organic material, where collective coupling causes the polaritonic states to be delocalized. The experiments show the potential for increasing carrier transport by polaritonic means: One could imagine building a transistor in which an external modulation of the cavity vacuum electric field would control the current.

Symmetry and beyond

The examples in this article make clear the enormous potential to modify and control material properties—even ground-state ones—by inducing hybrid light–matter states. Still, despite the simplicity of the coupling process, many of the underlying fundamental changes associated with strong coupling remain poorly understood. Experiments suggest the whole behavior of a coupled system switches, as if it has formed a new state of matter driven by the redefinition of its energy levels.

In the extreme case of USC, a forbidden bandgap appears and all the energy levels are shifted. Vibrational USC provides a practical way to generate polaritonic states with smaller linewidths than those of either the bare optical transition or the molecular vibration, which indicates longer-lived hybrid states. The dynamics of bond breaking in the ground state could therefore be radically modified, yet the potential impact of USC on chemistry remains to be explored.

The impact of symmetry in strongly coupled solid-state systems has yet to be examined. Beyond modifying transport properties, strong coupling also appears to be a promising route for controlling phase transitions—not just by tuning familiar ones, such as in superconductivity, but also by inducing new ones. For example, Yuto Ashida (at the University of Tokyo) and collaborators recently proposed a method for turning a paraelectric material into a ferroelectric one.[17]

In the terahertz regime, weak interactions such as intermolecular forces strongly influence the macroscopic properties of many materials and molecular systems. Strong coupling in those systems may open the door to controlling and enforcing new self-assembly processes.[18] It could also act on biological architectures and affect their functions. Chirality is an important property in those cases, and it needs to be explored in the context of strong coupling, material properties, and chemical reactivity.

Controlling material properties with polaritonic states is an unusually multidisciplinary endeavor, which explains the excitement and broad interest it has recently generated and the new perspectives it has opened. There will no doubt be many more surprises.

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