# Optical Control
# through Light
# Transmission

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The simple action of passing laser light through an optically transparent system may enable researchers to control a number of mechanical and optoelectronic processes. These novel interactions of light present an array of useful applications in optical switching, optical binding and fluorescence imaging.

W hen resonant laser light interacts with matter, it commonly undergoes gains or losses by exchanging energy with the atomic or molecular components of the material. However, a beam for which the material is optically transparent can engage in interactions in which transmitted light suffers no measurable change. In this sense, the beam is passive—yet the material may still exhibit an active optical response: The light-matter couplings that ensue can produce extensive mechanical or optoelectronic effects.

For example, couplings of this type support the widely exploited mechanical effects of "optical tweezers," which, along with other forms of particle trapping and manipulation, depend on positioning. Motion toward any configuration of a local energy minimum is impelled by optically induced forces and torques.

A familiar example of an optoelectronic effect associated with a passive beam is the optical Kerr effect. Here, the phenomenon exhibits the capacity of throughput light to produce a change in refractive index within a stimulus beam, modifying the polarization character of a secondary transmitted beam.

In addition to these well-known effects, a range of other, less familiar phenomena also involve throughput beam
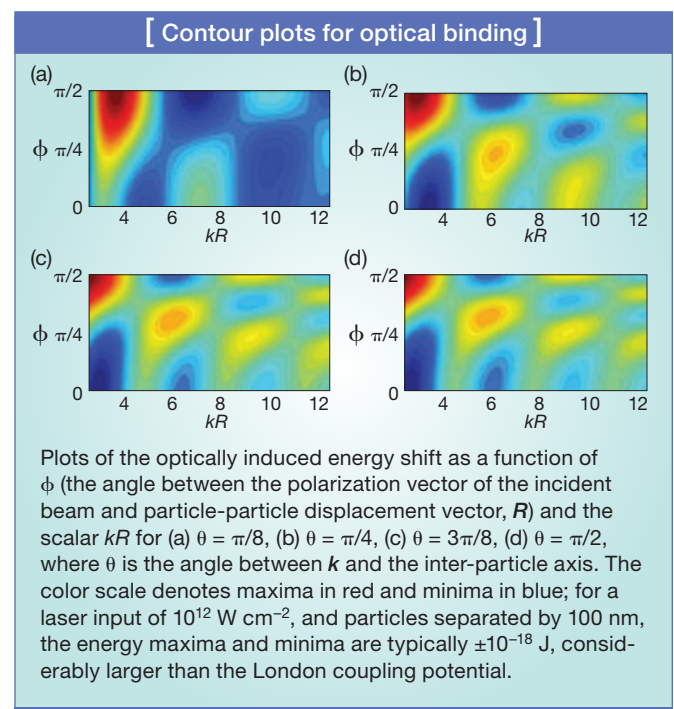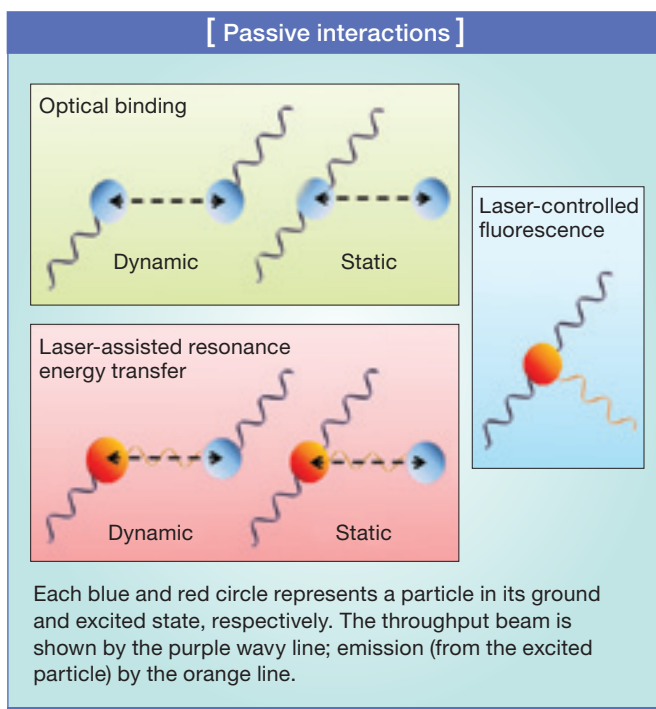
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interactions. The first to be experimentally realized was an effect known as "optical binding"—in which propagating laser light coherently modifies the form and magnitude of inter-particle coupling. This rapidly expanding field arose from earlier studies based on quantum electrodynamics (QED).

Recently, researchers have begun to explore several other fundamentally related processes that are also amenable to experimental investigation. These include the optical control of the flow of energy within complex heterostructures and the control of fluorescence emission rates.

## Optical binding

The nanoscale attraction between electrically neutral particles is primarily determined by their dynamic electronic properties. The best known interaction of this type, known as the London interaction, is characterized by an inverse sixth-power dependence on distance. Such forms of coupling operate in systems that range from free atoms to the composite particles of ceramic materials. The phenomenon of optical binding produces a modification to such interactions that arises from inter-particle coupling with electromagnetic radiation. The induced pair-wise interactions of optical binding give rise to laser-dependent forces and torques (although the forces are not necessarily attractive in form).

[ **Passive interactions** ]

Optical binding

Dynamic          Static

Laser-controlled fluorescence

Laser-assisted resonance energy transfer

Dynamic          Static

Each blue and red circle represents a particle in its ground and excited state, respectively. The throughput beam is shown by the purple wavy line; emission (from the excited particle) by the orange line.

[ **Contour plots for optical binding** ]

(a) $\pi/2$   $\phi$ $\pi/4$   0   4   6   8   10   12   $kR$

(b) $\pi/2$   $\phi$ $\pi/4$   0   4   6   8   10   12   $kR$

(c) $\pi/2$   $\phi$ $\pi/4$   0   4   6   8   10   12   $kR$

(d) $\pi/2$   $\phi$ $\pi/4$   0   4   6   8   10   12   $kR$

Plots of the optically induced energy shift as a function of $\phi$ (the angle between the polarization vector of the incident beam and particle-particle displacement vector, **R**) and the scalar $kR$ for (a) $\theta = \pi/8$, (b) $\theta = \pi/4$, (c) $\theta = 3\pi/8$, (d) $\theta = \pi/2$, where $\theta$ is the angle between **k** and the inter-particle axis. The color scale denotes maxima in red and minima in blue; for a laser input of $10^{12}$ W cm$^{-2}$, and particles separated by 100 nm, the energy maxima and minima are typically $\pm 10^{-18}$ J, considerably larger than the London coupling potential.

When two or more beams are involved, these optical binding forces can override local optical tweezer effects, imposing a different, competing structure. Researchers have experimentally observed many optically induced arrays, and the phenomenon is now advocated as a tool for the optical manipulation and configuration of particles. Similar effects have been reported in optically induced forces between parallel waveguides.

In QED calculations, a passive beam is represented as undergoing stimulated reemission by particle pairs. Accounting for concurrent dynamic and static mechanisms, one can determine a general expression for the optically induced energy shift. The full expression can be used to produce contour plots that represent the variation of potential energy with respect to the positions and orientations of each particle.

The results take the form of energy landscapes that exhibit highly detailed topographic features. Analysis of such energy surfaces facilitates the determination of the stability points associated with optical binding, and the identification of local forces and torques. Each pair potential provides a template for the optical assembly of larger numbers of particles, providing blueprints to optically fabricate structures with nanoscale dimensions, or to optically modify the forces that operate in nanoparticle self-assembly.

## Optical control of energy transfer

The mechanical effect of optical binding is a parametric phenomenon, signifying that the states of the light beam and the particles emerge unchanged. A more recently discovered but closely related nonparametric process differs in that the optical engagement involves radiationless resonance energy transfer (RET). Before we can explain the laser-controlled effect, it is critical to first understand the nature of RET.

In RET, real (rather than virtual) electronic transitions are involved, and our description is accordingly cast in terms of discrete molecules—the theory applies equally to atoms, ions or chromophores. In its spontaneous form, RET, which commonly follows light absorption by a donor molecule, enables the translation of associated electronic excitation to an acceptor molecule—a process that is crucial to the mechanisms of light-harvesting complexes. QED calculations have shown that radiationless and radiative energy transfer are the short- and long-range asymptotes of a single unified mechanism.

In the laser-controlled effect, when one applies the passive laser beam, energy transfer may be enhanced by conferring an optical response known as laser-assisted resonance energy transfer. The efficiency of this process scales linearly with laser intensity. Comparing the results with optical binding reveals a close relationship between the parametric and nonparametric couplings, since both involve elastic forward-scattering of the passive beam without any net absorption or stimulated emission. Yet, despite this similarity in description, laser-assisted RET has quite different applications.



**[ A device for all-optical switching ]**
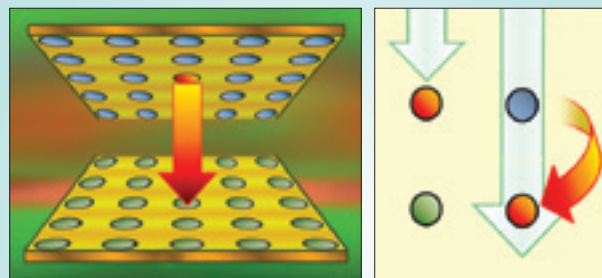
A two-dimensional square-lattice configuration for an all-optical switching device: (Left) side-view (not showing spacer layer between the two arrays); the initial excitation (red dot) is transferred only to its partner directly underneath; (Center) with the passive beam yet to enter, there is no transfer of excitation; and (Right) with the beam passing through the system, transfer occurs.



**[ Laser-enhanced near-field emission ]**
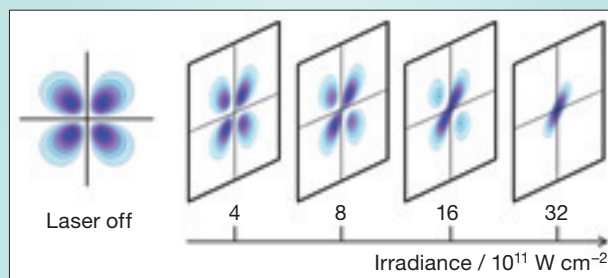
Representative contour plots of the laser-enhanced near field detected by a probe with transition dipole perpendicular to the source. With typical values for the optical wavelength, beam width and energy level spacings, these plots will apply to a planar area of $200 \times 200$ nm square, located 40 nm from the source.

For example, having the ability to exert optical control over the migration of energy provides a basis for all-optical switching. In an adapted form of laser-assisted RET, the spontaneous process is completely disabled. This can be achieved in an arrangement where the vector displacement of the acceptor from the donor—and each of their transition dipoles—are all mutually perpendicular; spontaneous RET is then excluded by geometry.

Alternatively, spontaneous transfer will be inhibited when the electronic transition in either the donor or acceptor is one-photon-forbidden but two-photon-allowed. In either case, the switching action induced by the transport of energy is activated only when the passive beam is "on." The characteristics of near-field emission by the donor are also modified, particularly in a heightened directionality that is more commonly associated with far-field, propagating light.

Modeling has indicated that an efficient, all-optical heterostructure device built on a two-dimensional square-lattice configuration can operate with readily achievable laser irradiances in the range $10^{10}$-$10^{12}$ W cm$^{-2}$. The parallel-processing capability of such a device introduces a variety of applications. For example, pixel-based images, which are written by donor excitation, can be controllably transferred with high fidelity to an acceptor film. In the realm of optical communications, possibilities arise for the system to act as an ultrafast information buffer; "slow-light" methods are also being pursued for similar aims.

## Laser-controlled fluorescence

Apart from two-center interactions, passive laser control can also influence a radiative effect that commonly competes with energy transfer in fluorescent molecules. Generally such molecules have two competing means of excited state decay—RET and fluorescence. Whether only one or both decay pathways are significant, the passage of a moderately intense passive laser beam may enable the optical control of the decay lifetimes, as the excited-state lifetime, $\tau$, is simply expressible as follows: $\tau^{-1} = \tau^{-1}_{FL} + \tau^{-1}_{RET} + KI$. The first two terms on the right side of the equation are well known; they correspond to excited-state lifetimes due to fluorescence and spontaneous RET, respectively. The newly discovered third

term exhibits proportionality to $I$, the intensity of the passive beam. Quantum electrodynamics shows that the proportionality constant $K$ expresses a dependence on both laser-controlled fluorescence and laser-assisted RET.

Suppose that, in a certain sample, we have a single fluorescing molecule with no suitable acceptor for the released photon energy. Transmission through the system of a passive beam can characteristically modify the intensity of emission, and its associated optical polarization and temporal behavior. The mechanistic conditions are entirely different from those that would operate under the delivery of resonant radiation, which would promote stimulated emission. Here, no net absorption or stimulated emission occurs since the throughput beam emerges unchanged.
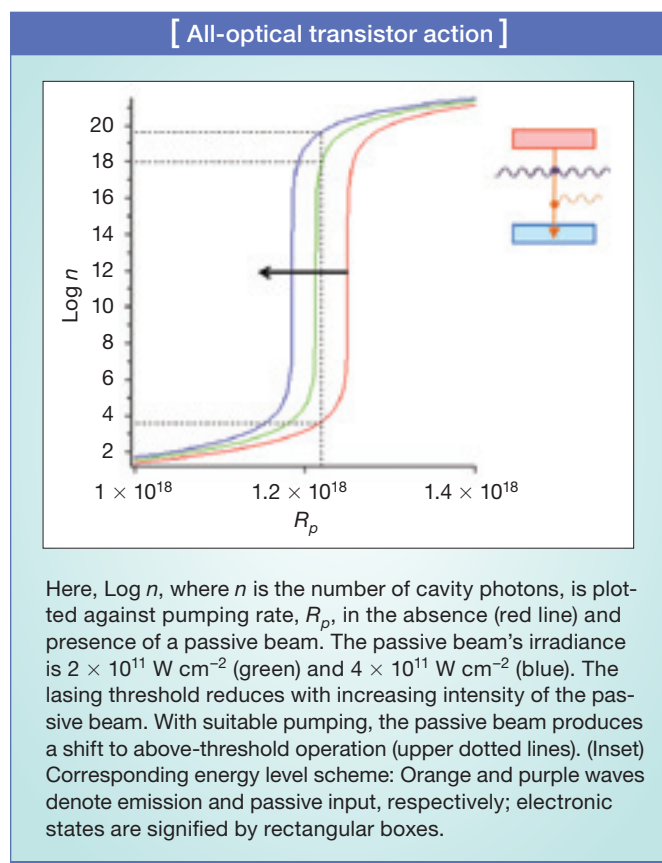
Specifically, elastic forward-scattering of the off-resonant light couples with the fluorescence emission; the probe laser essentially confers optical nonlinearity on the emission process. It is interesting to reflect that a strong local static field, which is often associated with a metallic surface, may also significantly modify fluorescent decay. The all-optical mechanism can produce such modifications to spontaneous fluorescence, without any such surface, through direct interaction with the oscillatory field of the throughput passive radiation.

According to the latest calculations, fluorescence lifetimes will be reduced by about 20 percent for an input laser irradiance of $2 \times 10^{11}$ W cm$^{-2}$, and proportionately more for higher intensities; a maximum is imposed by the pumping rate of the laser that initially excites the system. Even at $10^{10}$ W cm$^{-2}$, this change in fluorescence lifetime will be readily measurable with modulation detection techniques. One of the most commonly measured structural parameters, the degree of fluorescence anisotropy, which measures the difference in fluorescence intensities through polarizers parallel and perpendicular to the polarization of an excitation beam, is also subjected to a characteristic change due to the interaction with the probe beam.

## Optical transistors

Looking to the future, there is another tantalizing prospect associated with laser-controlled fluorescence. Research has recently shown that the output of a three-level laser, optically pumped just below threshold, may also be strongly enhanced on an ultrafast timescale through laser-controlled fluorescence. Analysis demonstrates that pulses of light from a passive laser, acting as an off-resonant input, modify the amplification kinetics of the active medium and thereby enhance laser output. The results of calculations based on QED for a representative three-level system highlight a significant potential for all-optical transistor action.

For a constant pumping rate, at a level indicated by the dotted vertical line (see figure to the left), the system operates below threshold with no input laser signal. Introducing a passive beam with an irradiance approaching $2 \times 10^{11}$ W cm$^{-2}$, the output climbs by approximately 14 orders of magnitude, rising

[ All-optical transistor action ]



Here, Log $n$, where $n$ is the number of cavity photons, is plotted against pumping rate, $R_p$, in the absence (red line) and presence of a passive beam. The passive beam's irradiance is $2 \times 10^{11}$ W cm$^{-2}$ (green) and $4 \times 10^{11}$ W cm$^{-2}$ (blue). The lasing threshold reduces with increasing intensity of the passive beam. With suitable pumping, the passive beam produces a shift to above-threshold operation (upper dotted lines). (Inset) Corresponding energy level scheme: Orange and purple waves denote emission and passive input, respectively; electronic states are signified by rectangular boxes.

to 16 orders if the input is doubled—typical of a laser operating at threshold. Transistor action with respect to the passive beam is clearly exhibited. The beauty of this scheme is that it is not limited to an operation with any specific material; the challenge is now to find three-level systems optimizing the necessary nonlinear optical characteristics.

## Applications

It is remarkable that each of these widely varying phenomena owes its origin to a fundamentally similar form of electromagnetic interaction, namely the stimulated reemission of passive, off-resonant transmitted light. Indeed, it is only through their common foundation in quantum electrodynamics that any of these processes has been discovered. Nonetheless, very different applications are beginning to emerge.

The experimentally well-established process of optical binding offers the prospect of implementation in a variety of applications that involve optically controlled self-assembly. Suitable tailored laser fields can manipulate and pattern micro- and nano-particles, forming a range of predesigned structures. Many researchers have become interested in developing templates for the nucleation of meta-crystals with a variety of lattice structures; these are created by analyzing the pair energy landscapes. Such an approach is not restricted to colloids; when applied to biological cells, it opens the door to the development of new optical methods for tissue generation and cell differentiation. We may be able to create tools for studies across the natural sciences by exploiting the opportunities for self-organization based on optical binding with multiplexed laser fields.

The optical control of RET affords quite different applications. One of the most promising prospects is all-optical switching implemented in a synthetic heterostructure. To build a switching device, researchers could exploit recent advances in the construction of quantum dot arrays, with juxtaposed arrays sandwiching an optically transparent spacer layer. Technical realizations will determine the most suitable nanofabrication method, such as dip-pen nanolithography. One aspect currently under exploration is the sensitivity to transition dipole disorder.

Compared to most other nanoscale schemes, optically controlled RET is attractive for a number of reasons. It has the positive characteristics of viable operation at short UV/visible wavelengths without requiring expensive, nonstandard optical elements; above all, it offers a high-information-density and ultrafast response with high repetition rate and high efficiency.

We can also expect researchers to exploit newly discovered means of optically controlling fluorescence. When used as an analytical tool with a tunable laser source, wavelength

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variation of the passive radiation offers an additional dimension to sample interrogation; it helps to enable the differentiation of samples that appear indistinguishable by conventional optical methods.

In addition, nonintrusive, generically applicable laser methods can be used to secure better spatial resolution in fluorescence imaging applications, allowing the characterization of high-grade materials such as microcrystalline polymorphs. In the life sciences, methods that afford a combination of high spatial and temporal resolution are well suited to research on the dynamics of cell processes, as already well established in current multiphoton excitation techniques. Exploiting passively induced optical nonlinearity offers wider applications with significantly less expensive instrumentation.

We anticipate that the initial work on optical transistor action, based on the passive control of fluorescence, may also break ground for the broader development of new, all-optical methods to control the basic processes of photon absorption and emission. Such opportunities are not limited to fundamental applications; since operation does not require atomic motion and switching times are comparable to an excited state decay, there are clear advantages over the current generation of molecular switches—devices based on laser-activated emission will be much faster in operation and cycle time. In the ongoing progress of optoelectronics, a new generation of molecular devices will emerge, tailored for use in optical communications and information technology. ⏶

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