most common pathway for the formation of ammonium nitrate in a relatively clean atmosphere keeps the compound near chemical equilibrium with gaseous ammonia and nitric acid. As a result, that mechanism is unlikely to increase the number density of particles unless the atmosphere becomes supersaturated with nitric acid and ammonia.

To investigate what conditions may drive supersaturation and new particle formation in an urban setting, Donahue and his colleagues studied mixtures of common pollutants in the CLOUD (Cosmics Leaving Outdoor Droplets) chamber at CERN (figure 3). The facility—modeled after cloud chambers used in particle physics in the 1950s—provides an environment in which to simulate vapor and particle interactions under conditions representative of the atmosphere from the lower troposphere up to the stratosphere. UV light generated by a built-in fiber-optic system mimics the energy provided by the Sun's cosmic rays. That radiation ultimately controls how molecules ionize at different altitudes.

In a series of experiments designed to replicate winter conditions in polluted cities, the researchers introduced various combinations of nitric acid, ammonia, sulfuric acid, and their precursor molecules into the CLOUD chamber. They exposed the gases to a range of temperatures typical of the lower atmosphere and varied the radiation input to control ion distribution. Mass spectrometers and particle counters that encircle the chamber determined the resulting concentration of gases and the properties of newly formed particles.

The team found that at temperatures warmer than –15 °C, ammonium nitrate and sulfuric acid followed a known pathway to form small clusters of molecules.[3] Once clusters of ammonium sulfate reached a threshold size, ammonium nitrate began to condense onto the clusters, resulting in rapid particle growth—above 100 nm/hr—as long as the temperature remained below 5 °C. At temperatures colder than –15 °C, which can occur in humid air outflows above some clouds, the nitric acid and ammonia gases nucleated through an acid–base stabilization mechanism to form particles of ammonium nitrate, which then grew via additional condensation of ammonium nitrate.

Calculations showed that the critical size at which ammonium nitrate begins rapid growth depends on the maximum concentrations of the ammonia and nitric acid.[4] Once that size is reached, rapid growth continues until the precursor vapors return to the chemical equilibrium commonly found between ammonia, nitric acid, and ammonium nitrate.

Large-scale atmospheric transport models do not take into account the newly proposed path to particle formation. The CLOUD experiments suggest that it may be important instead to consider those vapors using dynamic calculations. The researchers propose that variations in temperature and emission sources in cold urban settings provide the conditions necessary for localized supersaturation of nitric acid and ammonia. By imposing regulations to control nitrogen emissions, urban planners could reduce the concentration and growth of particles and improve air quality.

**Rachel Berkowitz**

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# Chip-scale sensor detects light's orbital angular momentum

The photocurrent in an unusual material could facilitate an increase in the information density of optical communications.

Light often has spin angular momentum, more commonly referred to as left or right circular polarization. Only in the past 30 years have researchers been able to impart orbital angular momentum (OAM; see the article by Miles Padgett, Johannes Courtial, and Les Allen, PHYSICS TODAY, May 2004, page 35). Instead of the usual flat wavefront, light with nonzero OAM has a helical wavefront that turns like a corkscrew in the direction of propagation. The quantum number $m$ describes how tight the light's corkscrew motion is. For example, $m=1$ means a full clockwise rotation in a wavelength, and $m=-2$ means two full counterclockwise rotations in a wavelength.

OAM provides additional degrees of freedom for encoding information in optical communications. Modern optical networks rely on wavelength and intensity modulations to send data; the addition of polarization and OAM could pack in more information without boosting traffic. What's more, unlike polarization, which has two modes, $m$ can take any integer value. Modes of different $m$ are also orthogonal and wouldn't interfere with one another during transmission.

Ordinarily, measuring OAM requires a roomful of bulky optics. But Ritesh Agarwal at the University of Pennsylvania and his colleagues have found a compact way to detect light's OAM through the induced photocurrent in a device that's just tens of micrometers across.[1] Their technique could make it easier to read out information encoded in the OAM.

## Gaining momentum

To measure $m$, researchers typically interfere light with a reference beam or send it through a hologram or specially designed aperture. The nature of the interference or diffraction pattern manifests the OAM. But those measurements not only require table-scale setups; they are also limited in the number of modes they can measure. For example, most holograms can detect only one particular value of $m$, so a new one must be swapped in for each mode.

In CCDs and other solid-state sensors, light is detected through its interactions with matter. A material's response depends primarily on the local incident number of photons—that is, the intensity. Phase information is usually lost when intensity is converted to photocurrent. Because a clockwise helical

wavefront and a counterclockwise one have the same donut-shaped intensity profile, a phase-sensitive measurement is essential for characterizing light with OAM without introducing holograms and apertures.

Agarwal has been contemplating such a measurement for five years. Back in 2015 he and his team found a way to detect a light beam's circular polarization—its spin angular momentum—with a silicon nanowire.[2] They were interested in extending the capability to OAM but weren't sure how to do it. A couple years later, a breakthrough came when the team was trying to understand the properties of a new group of materials, Weyl semimetals.

## Mean-Weyl . . .

Identified for the first time in 2015, Weyl semimetals have a broken symmetry, generally inversion symmetry, that allows them to host quasiparticles that behave as Weyl fermions—massless, charged, and chiral. (For more on Weyl semimetals, see PHYSICS TODAY, December 2019, page 24.) Agarwal and his colleagues found that the photocurrent in Weyl semimetals was sensitive not to the light intensity but to the intensity gradients.[3]

Photocurrents in most materials depend only on the light's local intensity. But when excited at a normal angle, a crystal with mirror symmetry reacts not to the local intensity but to how the local intensity compares with the surrounding intensities, or the intensity gradient. In that case, the photocurrent is nonlocal and results from an imbalance, in both real and momentum space, in the density of excited electrons. Mathematically, it is described by the terms beyond the dipole approximation in the multipole expansion of the light–matter interactions. The symmetry cancels out the dipole response, which would otherwise obscure the other terms. Although Weyl semimetals aren't the only materials with the requisite symmetry, they have the added advantage of a nonlinear optical response that's among the largest identified to date.

In their intensity-gradient experiment, the researchers measured a circular photocurrent that could be explained only by the nonuniform distribution of the light. But in addition to a nonuniform intensity, light with nonzero OAM has a circling phase gradient that depends on $m$. Could a photocurrent sense a phase gradient as well? The group's calculations indicated that the measurement should be possible.

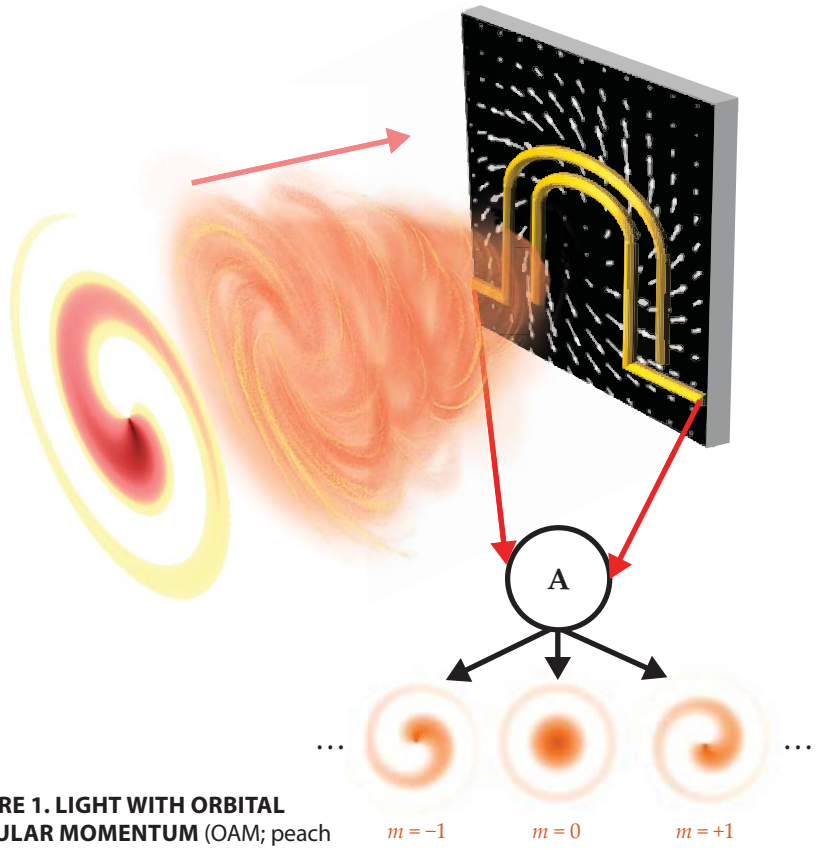


**FIGURE 1. LIGHT WITH ORBITAL ANGULAR MOMENTUM** (OAM; peach swirl on the left) excites photocurrents in a material (black rectangle). Usually, the photocurrent depends only on the light intensity, which doesn't vary with OAM. But in materials with the right symmetry, it arises in part from light's phase gradient (white arrows)—that is, the transfer of OAM from photon to electron. Using specially designed U-shaped electrodes (yellow), researchers measure the photocurrent with an ammeter (A) to determine the OAM quantum number $m$. (Adapted from ref. 1.)

## A new phase

Agarwal and his graduate student Zhurun Ji prepared a 50- to 200-nm-thick layer of the Weyl semimetal tungsten ditelluride and designed U-shaped electrodes, shown in figure 1, to probe the photocurrent generated by light with OAM. The electrode shape needs to distinguish the current due to the OAM phase gradient from the current due to intensity gradients and other effects. Although a few other electrode shapes work, the U shape is the best at measuring a radial current and unambiguously canceling out the other effects.

Before reaching the $WTe_2$ detector, a beam with OAM passes through a rotating quarter-wave plate that cycles the polarization from 0° linear to left circular to 90° linear to right circular and back to 0° linear. The measured radial photocurrent, shown in figure 2a for beams with $m$=+1 and −1, has three components: one that depends on the linear polarization state; one, $J_c$, that depends on the circular polarization state; and one that is independent of polarization and includes thermal currents. To distinguish between the effect of the intensity gradient and that of the phase gradient, the researchers exploit the components' differing dependencies by comparing beams with OAM $+m$ and $-m$, which have opposite helicities but the same intensity profiles.

The photocurrent is not the same for left and right circular polarization states (the left and right peaks in figure 2a). And the difference, arising from $J_c$, is around the same magnitude but swapped for

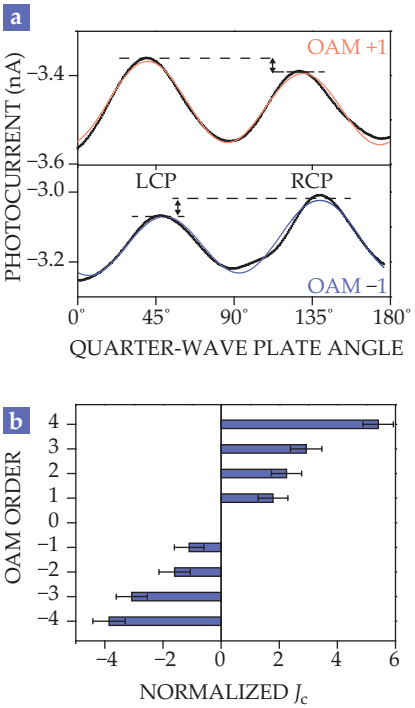


**FIGURE 2. A WEYL SEMIMETAL'S PHOTOCURRENT** is sensitive to light's orbital angular momentum (OAM). **(a)** Before reaching a thin layer of tungsten ditelluride, light passes through a rotating quarter-wave plate, which cycles the polarization from linear to left circularly polarized (LCP) to orthogonal linear to right circularly polarized (RCP) and back to the original polarization. Part of the current depends on circular polarization and leads to the difference in the left and right photocurrent peaks, which correspond to LCP and RCP. That difference is around the same magnitude but swapped for OAM +1 (upper curve) and −1 (lower curve). **(b)** The photocurrent component $J_c$ that varies with circular polarization is proportional to the magnitude and helicity of light's OAM. (Adapted from ref. 1.)

OAM +1 and −1. The researchers found the same behavior for all OAM modes that they measured. It turns out that $J_c$ corresponds to the photocurrent response from the phase gradient and grows proportionally with $|m|$, as shown in figure 2b for OAM up to ±4. It results from light transferring its OAM and energy simultaneously to the electrons in the material. Because the light's phase isn't uniform, there is once again a spatial imbalance of excited electrons and a net current either along or perpendicular to the gradient.

To complement the chip-scale detection of OAM, researchers have reduced the production of light with OAM to chip-scale too. Published back to back with Agarwal's paper, the University of Pennsylvania's Liang Feng and his colleagues report producing light with five different OAM values with the same microlaser.[4] In their scheme, light circling in a microscale semiconducting ring was scattered by ridges on the inner surface of the ring such that the light's circling became a helical propagation. Control over the OAM chirality came from two waveguide arms, one pumped in clockwise light and the other, counterclockwise.

**Heather M. Hill**

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