how electronic energy is converted into heat and work and for understanding photochemical processes such as photosynthesis, the mechanism of vision, electron transfer reactions, and the photostability of DNA.

Along conical intersections, small changes in molecular geometry can have dramatic effects on the electronic configuration of a molecule. These effects of the nuclear dynamics can be picked up by measuring the photoelectron spectrum in a time-resolved experiment in which a short-duration laser pulse excites the molecule (called the pump pulse) and, after a short time delay, a second probe pulse ionizes it. However, the results are ambiguous in terms of which electronic state emitted the photoelectron (*4*, *5*).

Wörner *et al*. (*1*) attacked this ambiguity by investigating the dynamical rearrangement of the electrons directly as a molecule moves through a conical intersection by measuring the high-harmonic spectrum (HHS) induced when an electronically excited molecule interacts with an intense laser field. High-harmonic generation, the production of radiation at many times the frequency of the driving ultrafast laser pulse, occurs in three stages. First, the intense electric field of the laser pulse removes an electron from an atom in a process known as field ionization. The free electron is then accelerated in the electric field of the laser. However, the electric field in a light pulse oscillates, so in half an optical cycle (1.3 femtoseconds for a pulse centered at 800 nm in the near infrared), the sign of the field reverses. The electron is accelerated back toward its source, where the resulting collision releases energy and generates light at harmonics of the driving laser field.

The relative intensity of the HHS of molecules is sensitive to their electronic structure, and this property has been used to reconstruct the electronic wave function of the highest occupied molecular orbital in some simple molecules (*6*). The interpretation of this experiment has been controversial for two reasons. First, molecules other than H_2^+ have many electrons, and the concept of a "one-electron" wave function is merely a theoretical construction to make electronic structure calculations easier to handle in a computer. Second, a fundamental tenet of quantum mechanics is that the wave function, which is usually described by complex numbers, is not in itself an observable quantity (*7*). This debate was resolved when it was realized that the observed HHS resulted from a coherent interaction between the ionized photoelectron with the hole (the empty level) it left behind, and so the experiment on a ground state with a single configuration is largely blind to the configuration interactions in the true, many-electron wave function (*8*, *9*).

This framework is important for interpreting the results of Wörner *et al*. (*1*). They used laser pulses to pump a molecule, nitrogen dioxide (NO_2), into an excited electronic state before observing the HHS with a second intense pulse. They created a spatial distribution of excited-state molecules by creating an interference pattern with two identical pump pulses, which allowed them to separate the spectrum of ground-state molecules from the electronically excited ones. The interesting observation is that the HHS of the excited state is sensitive to the time delay between the pump field and the intense probe pulse that created the HHS.

Wörner *et al*. (*1*) argued that this time dependence reflects the electronic dynamics that results when the bending-mode vibrational state created after electronic excitation moves along the excited-state surface and reaches a geometry where it intersects with the ground-state surface (see the figure). Because the electronic configuration in the electronically excited state is more easily ionized than the ground-state configuration, a modulation was observed with a frequency characteristic of this vibrational frequency on the excited-state surface. Although the nuclear motion across a conical intersection has been observed before (*4*, *5*), this study reports the first direct observation of the dynamic rearrangement of electronic configuration in the vicinity of a conical intersection. Thus, the use of extremely fast light pulses—on close to attosecond time scales—has allowed the observation of events triggered by electronic transitions in molecules and shone light on the description of their electronic structure.

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ECOLOGY

Grass Trumps Trees with Fire

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Feedbacks between rainfall, fire, and vegetation govern transitions between forests, savannas, and grasslands.

Ecologists have long assumed that forests, savannas, and grasslands change gradually over space and time, with tree cover responding linearly to gradients in precipitation, aridity, fire disturbance, and grazing pressure. However, a growing body of evidence suggests that these biomes are self-reinforcing and that transitions between them can be nonlinear, governed by feedbacks at local and regional scales (*1*–*3*). Two reports in this issue, by Staver *et al*. on page xxxx (*4*) and by Hirota *et al*. on page yyyy (*5*), find evidence for these feedbacks and transitions at the global scale. These results suggest that global climate change will be substantially influenced by nonlinear behaviors and feedbacks between biophysical and human systems.

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Sudden transitions between forests, savannas, and grasslands, and the feedbacks that drive them, have been observed at local to continental scales (*6*, *7*). Sudden transitions at global scales were thought not to be observable, because local heterogeneity and small-scale transitions would appear as a more gradual change at larger scales (*8*).

Staver *et al*. used tree cover derived from satellite data, together with global data for annual and seasonal fire frequency and precipitation, to examine whether forests and savannas are alternative stable states and, if so, which mechanisms maintain these states.

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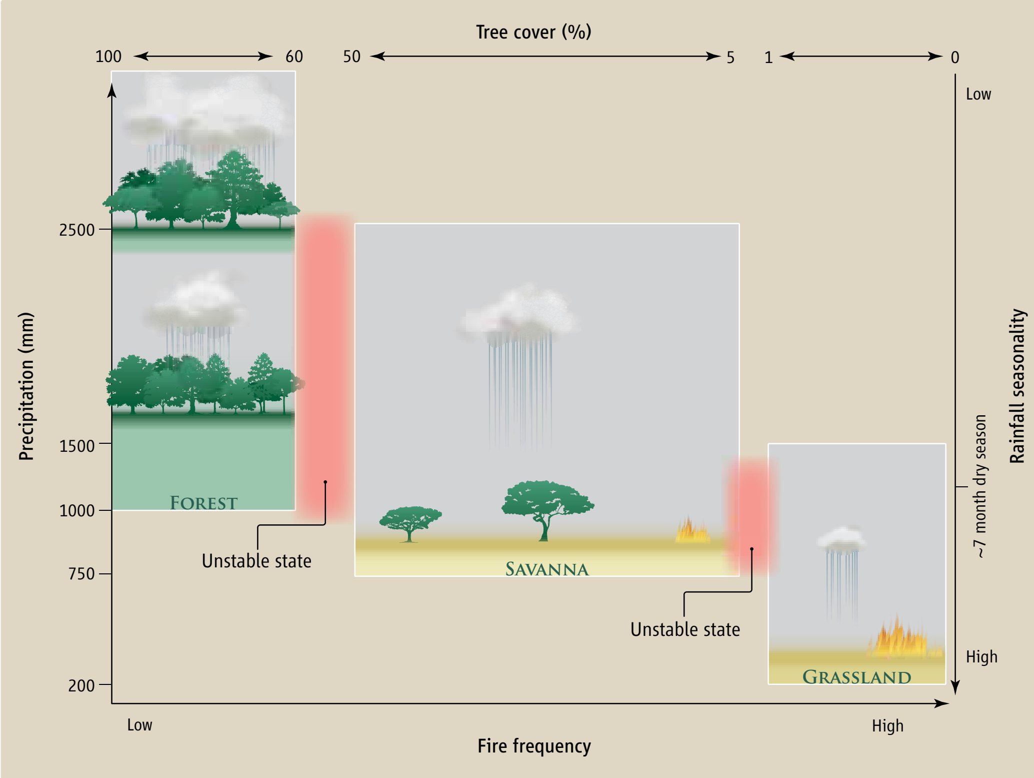

Transitions between biomes. Staver *et al*. (*4*) and Hirota *et al*. (*5*) have analyzed global data sets to identify transitions between biomes. The results show that areas with regular rainfall above 2500 mm/year are forest (>60% tree cover). Areas receiving rainfall between 1300 and 2500 mm/year can persist in either a forest or savanna state, depending upon the strength of the fire-grass feedback (*4*, *5*). Tree cover between 50 and 60% is an unstable state and demarcates forest from savanna. Areas receiving highly seasonal rainfall between ~600 and 1300 mm/year can persist as either savanna or grassland, depending on fire frequency (*5*, *9*). The effects of grazing and hu man intervention are not shown.

They excluded high-elevation areas, areas with high human impacts, and areas where most rainfall occurs during the nongrowing season. A forested state (more than 60% tree cover) was predominant in areas receiving more than 2500mm/year rainfall and where rainfall was temporally uniform (see the figure). Areas receiving rainfall between 1000 and 2500 mm/year were either forest or savanna, but few areas supported tree cover between 50 and 60%. Savannas existed where rainfall was highly seasonal and the fire–grass feedback was strong. In Australia, extreme rainfall seasonality precluded forests even in areas receiving up to 2500 mm/year of rain.

Hirota *et al*. used the same tree-cover data as Staver *et al*., but with different precipitation data. They only excluded parts of the Saharan and Australian deserts. By relating tree cover to mean annual precipitation, the authors identified three normal distributions that were best explained if forests, savannas, and treeless models were included. Thresholds were sharp between these states, with forests (>60% cover) found mainly above 2500 mm/year rainfall, savannas (5 to 50% tree cover) most common between 750 and 2000 mm/year rainfall, and treeless areas dominating below 750 mm/year rainfall. The extreme rarity of areas with intermediate tree cover suggests that transitions between these biomes are rapid.

Adding these global results to previous studies at local and continental scales (*3*, *6*, *9–11*) presents a strong argument that forests, savannas, and grasslands are alternative stable states at the global scale, maintained by three main mechanisms: a strong feedback between vegetation and precipitation; a strong feedback between rainfall seasonality and grass; and a very strong feedback between grass and fire (see the figure).

Both reports identify an unstable state at 50 to 60% tree cover; either trees take hold and promote their own growth hydrologically (and suppress fire), or grasses take hold and promote their expansion through fire. This work has implications for the resilience of these biomes in the Southern Hemisphere. Most notably, large areas of savanna in Africa could shift to forest (if fire and grazing are suppressed), and large areas of forest in South America could convert to savanna [although degraded forest does not necessarily transition to savanna (*11*)], as climate change and local human impacts such as logging interact with rainfall seasonality and fire.

The two reports show convincingly that forests, savannas, and grasslands are distributed discontinuously at the global scale, but the authors do not analyze several important mechanisms. Large herbivores (such as horses and antelope) evolved in concert with savannas and grasslands (*12*), and their feedbacks with grasses are known (*3*, *4*, *9*). Topography may also influence microclimates and thus fire spread and vegetation (*9*). Finally, prehistoric and historic human activities had a sizable influence on the forests, savannas, and grasslands that exist today (*13*).

Humans will continue to influence the distributions and resilience of these ecosystems on multiple scales; fire suppression, grazing of domesticated animals, forest harvests, restoration efforts, and contributions to climate change all have effects (*5*, *14*). Future studies should examine the universality of the feedbacks driving biome transitions in both hemispheres, as well as the impacts of human activities on these feedbacks, to assist the development of better-informed management and restoration plans.

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