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Teaser: New theoretical calculations show that an x-ray free-electron-laser pulse can generate transitions in excited nuclei via an indirect process involving electron capture.

Since they became available to users in 2010 [1], x-ray free electron lasers (XFELs) have been transformational tools for many scientific disciplines. These kilometer-sized machines generate x-rays with unprecedented brightness and pulse durations down to a few femtoseconds (10^{-15} s). It is not always entirely clear how these bright pulses interact with targets. In the case of nuclear physics, the expectation was that XFEL light would predominantly interact with nuclei through direct photoexcitation. But a new theoretical study of excited nuclear states has revealed that, surprisingly, XFEL-nuclear interactions are dominated by a secondary mechanism. As described in *Physical Review Letters* [2], this indirect photo-excitation mechanism – which involves electron capture from the laser-induced plasma – is over a million times more efficient. The results may be interesting for future energy storage applications, in which excited nuclei perform as nuclear batteries.

Free electron lasers are a big step up in x-ray generation, delivering 10^8 more photons per volume than synchrotron sources. The remarkable performance of XFELs is achieved by accelerating high-quality, ultrashort electron bunches to GeV energies in a linear particle accelerator. Radiation is emitted from the periodic transverse oscillations of the electron bunch passing through an undulator, a periodic arrangement of magnets. The radiation interacts with the electron bunch, which yields density modulations on the bunch. This density modulation enhances the x-ray emission process, and lasing can occur. There are currently three operational XFELs in the world: the LCLS in the U.S., SACLA in Japan, and FLASH in Germany, with a prolific scientific production and ever increasing experimental demands and number of users. Seminal achievements include the creation of some of the hottest plasmas on earth [3], the first atomic inner-shell x-ray laser [4], and structural studies of proteins with crystallography [5]. In these examples, x-rays play a role in understanding atomic and molecular processes.

While they hold the promise to revolutionize atomic physics and structural biology, XFELs could also help achieve breakthroughs in nuclear physics. Jonas Gunst of the Max Planck Institute for Nuclear Physics in Heidelberg, Germany, and his colleagues report the unexpected result that XFELs can indirectly trigger nuclear excitation processes in nuclear isomers, by coupling nuclei to the atomic shell [2]. Nuclear “isomers” [6], a term borrowed from chemistry, are excited states of a nucleus that have the same number of

neutrons and protons (as opposed to isotopes having different number of neutrons). Typical half-lives of nuclear excited states are on the order of 1 picosecond, but some of them can last several hours. According to the findings of Gunst *et al.*, triggering these excited states to release their energy on demand seems within reach. The authors describe the interaction of an XFEL with one of the isomeric states of molybdenum, ^{93m}Mo , which has an excitation energy of 2.5 MeV and a lifetime of nearly 7 hours. Nuclear decay of the isomeric level can be enhanced by exciting the nucleus to a less stable, intermediate level lying above the isomer. X-rays from the XFEL can perform this transition via two different mechanisms.

The first route is the direct one: if the XFEL is tuned to 4.85 keV, then the x-rays can excite the nuclei resonantly to a higher excitation state, from which they rapidly decay back to the ground state, releasing x-rays and gamma rays. This direct photoexcitation process is, however, rather inefficient due to the fact that the energy width of the nuclear transition is 8 orders of magnitude smaller than the width of the XFEL spectrum. Therefore, only a small fraction of the laser photons are resonant with the nuclear transition. The second possible route for releasing the isomer's energy is indirect, so it was thought to be less important. This secondary mechanism, called nuclear excitation by electron capture, or NEEC, is a resonant process, in which the XFEL light removes electrons from the target atoms (in the isomeric excited state), and these free electrons form a plasma with a thermal energy of about a few 100 eV. An electron from this plasma can subsequently be captured into an empty state of the atomic shell (left vacant from the interaction of the XFEL with the atom). This captured electron gives its excess energy to excite a nuclear state. The NEEC process, first described in 1976 [7] and considered as a possible nuclear decay mechanism in stars, has so far not been observed in the laboratory (with synchrotron or Bremsstrahlung radiation). Observing this phenomenon will presumably require the photon density and energy from an XFEL, able to ionize enough electrons from the target to create the plasma and bring it to a high temperature.

Assuming an XFEL source of photons, Gunst *et al.* calculate that indirect photoexcitation through the NEEC process is at least six orders of magnitude more efficient than direct photoexcitation. One reason for this large discrepancy is that – unlike the direct photoexcitation of the 4.85 keV transition – the NEEC mechanism is less sensitive to the resonance condition, since the photons have a wide range of inner-shell electrons with which to interact. Moreover, the plasma lasts much longer than the XFEL pulse, so there is more time for electron capture to occur as opposed to direct photon-mediated excitation.

The consequences of this work could spark new life into the field of nuclear energy storage, where scientists have been studying for years the prospects of using metastable nuclear isomers in high-energy batteries. The advantage of isomers – which are typically produced in nuclear reactions or through irradiation – is that they provide nuclear power with no long-lasting nuclear waste. However, the challenge is the control of the release of the stored nuclear energy. Gunst *et al.* have shown that the light from an XFEL could

trigger the rapid decay of a nuclear isomer. Now, given that this process requires using a large and costly facility, it seems unlikely that there will be a practical energy-storage device in the near future, but this new work could stimulate research on other triggering schemes. Another application could be in the medical community. Long-lived nuclear isomers are of interest for a number of applications in nuclear medicine [8], where they can be used for therapeutic (endo-radiotherapy) or diagnostic (nuclear imaging) purposes. In the example of ^{93m}Mo -triggering described by Gunst *et al.*, a 1 MeV photon is emitted in the decay cascade from the triggering level to the ground state, and it can potentially be detected by modern imaging systems or target a specific tumor.

The authors are marching into a virtually virgin territory, as no existing x-ray source has yet enabled the experimental demonstration of NEEC triggering of isomer relaxation. This theoretical work should inspire experimental efforts at XFEL facilities that have sufficient photon energy and x-ray intensity to allow the NEEC process to occur. Researchers can isolate the NEEC signal by tuning their XFEL light out of resonance with direct photoexcitation, so that the only isomer emission should be from the indirect mechanism. The effort will be challenging, but the results could be a major breakthrough.

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Figure

About the Author: Félicie Albert is an experimental plasma physicist at the National Ignition Facility, Lawrence Livermore National Laboratory, who earned her PhD in Physics from the Ecole Polytechnique in France in 2007. She has conducted many experiments using high-intensity lasers in France and the U.S. Her areas of interest include the generation of novel sources of electrons, x-rays and gamma-rays through laser-plasma interaction and laser-wakefield acceleration. She is currently leading an effort to develop these sources for applications in high energy density sciences as well as doing experiments for the National Ignition Facility.