2.B Characterization of Laser-Generated X-Ray Sources for Nuclear Level Excitation

Gamma-Ray Lasers

Gamma-ray lasers have been the subject of many published proposals over the years, extensively reviewed by G. C. Baldwin *et al.*[1] More recently, the subject has received new vitality from the idea of nuclear-isomer excitation by x rays, following isomer generation by slow-neutron irradiation in a reactor.[2] The first step toward the realization of a γ-ray laser will most likely be the demonstration of nuclear isomer excitation with x-ray radiation. Such excitation depends on, and in turn contributes to, our knowledge of nuclear energy levels and their characteristics. The OMEGA laser system is capable of producing adequate x-ray flux levels for pumping certain nuclear candidates. An NLUF experiment to excite the thulium isotope ^{169}Tm on OMEGA has been proposed by C. B. Collins[3] of the University of Texas at Dallas (UTD) and is currently under way. Further experiments involving other nuclei are being planned jointly by LLE and the UTD group and are discussed here.

The principal obstacle to achieving x-ray lasing is that lifetimes of ionic levels decrease with Z (usually, as Z^{-4}). That requires the pumping power to scale as Z^6. On the other hand, nuclear-level lifetimes can be very long (up to years), when multipolarity changes by much more than 1, reducing the pumping requirements. In the past, neutron excitation was considered as a pump source for γ-ray lasers, but with discouraging results. This is mainly because neutron cross sections are small, of the order of 1 barn (10^{-24} cm^2), whereas photon cross sections are given by the Breit-Wigner formula $\sigma = (\lambda^2/2\pi)(\Gamma_r/\Gamma)$; for 1-Å radiation they typically are $10^{-19} - 10^{-17}$ cm^2. Here, Γ_r is the radiative width and Γ the total width of the upper level (this includes mostly internal conversion). This cross section for photon energy less than 100 keV can be several orders of magnitude higher than absorption losses, which in this range are mostly photoelectric (Compton scattering being small). Neutron fluxes required to cause *in situ* inversion must be unrealistically high. Also, intense bursts of neutrons are too energetic for cross sections to be sufficient, and neutron moderation is therefore necessary. The energy load on the moderator, however, will then have a destructive effect.

The lasing nuclei have to be part of a Mossbauer crystal; otherwise, the recoil energy causing shift and broadening of the γ photon will drastically reduce the stimulated emission cross section. If the crystal is perfect enough to exhibit the Borrmann effect, absorption losses in the Bragg direction will be drastically lower. Both the Mossbauer and Borrmann effects require minimum heating of the lasing medium by the bulk absorption of the pump radiation.

The procedure for the x-ray isomer pumping is as follows. First, isomeric nuclei are produced for a few months in a reactor by thermal-neutron capture in parent nuclei. Then, isotope and isomer separation produces a pure isomer sample of sufficient size – at least 1 cm^2 – to capture a sizable fraction of the x rays emitted by a laser target at a distance of about 5 cm. The thickness of the sample has to be the e-folding distance for photoelectric x-ray absorption at the pump wavelength. Any compromise on these sample preparation requirements will reduce the signal-to-noise ratio in detecting the nuclear decay following excitation by x rays, and will necessitate gathering statistics from many laser-target shots. Ideally, pumping by x rays should transfer the isomer-level population to a very short-lived level. In this case, the broad nuclear level will intercept a wider spectral band of pump x rays. This level should then decay to the upper γ-ray laser level, for which the lifetime should be longer than the laser pulse width (~ 1 ns).

The number of excited nuclei N can be simply estimated as[3]

$$N = F_1 \times F_2 \times F_3 \times F_4 \quad , \qquad (1)$$

where F_1 is the strength of the laser-irradiated x-ray source, in photons per keV per unit solid angle. F_2 is the nuclear level width (in keV) due only to the radiative transition. F_3 is the ratio of on-resonance cross section for both excitation and internal conversion, to the nonresonant (photoelectric) absorption cross section. Finally, F_4 is a correction factor that accounts for photon depletion around resonance. Only the ratio of cross sections appears because the x-ray line spectrum is continuous on the scale of the much narrower nuclear level width; thus, resonant absorption can take place away from line center, up to the point where resonant and nonresonant cross sections become equal. For the thulium experiment discussed below, $F_2 = 5.2 \times 10^{-13}$ keV, $F_3 = 560$, and $F_4 = 0.1$. Therefore, if $F_1 = 10^{14}$ keV^{-1}, the total signal is about 3×10^3 events, sufficient, when multiplied by the detector solid angle, to be observed with a small number of laser shots. This value of F_1 translates to an x-ray fluence of almost 10^{16} keV/keV into 2π solid angle (the thulium transition energy is 8.4 keV). A value of x-ray fluence above 10^{16} keV/keV is taken as a minimum for significant nuclear excitation of promising nuclear candidates. As shown below, OMEGA target shots can produce this and higher fluence levels on a single-shot basis for photon energies up to and beyond ~ 10 keV.

Proposed Nuclear Pumping Experiments

Three experiments have been designed for testing both the physical and instrumental aspects of nuclear pumping schemes. The nuclei involved are

(a) ^{169}Tm (thulium)
(b) ^{137}La (lanthanum)
(c) ^{110}Ag (silver)

The experiments will be taken up in this order, which is from least in cost to the most costly, but from the more difficult to the less difficult.

In the thulium experiment (see energy level scheme in Fig. 26.7), an excitation will be attempted from the nuclear ground level to the spin (3/2)+ level at 8.4099 keV. As an excitation source, a Cu^{+27} x-ray line from copper-coated targets was selected. The intense resonance line of this ion at 8.39 keV has been shown to be excited in OMEGA experiments (see below) with a spectral fluence of 10^{17} keV/keV. As shown above, this is sufficient to observe a signal of nuclear decay. The coin-

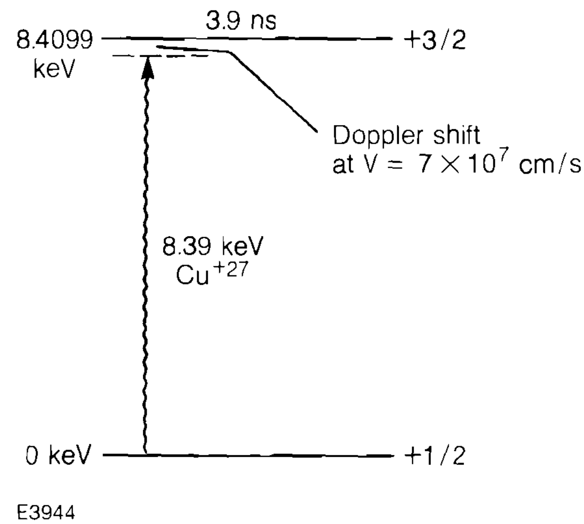

Fig. 26.7
Nuclear energy levels in ^{169}Tm. Excitation of this nuclear transition will be attempted with the 8.39-keV laser-produced x-ray line of Cu^{+27}.

cidence between atomic and nuclear transitions seems to be inexact, but the Doppler shift due to the plasma expansion from the laser-irradiated target could compensate for the energy difference.

In the geometry of the thulium experiment, the copper plasma expands toward the thulium sample, so that the Doppler energy shift would bring the two energies closer. An expansion velocity of 7×10^7 cm/s is required for perfect coincidence; this value is entirely typical of plasma expansion velocities, so an energy coincidence is almost certain. The spectrum of expansion velocities increases the probability of resonance with nuclear transitions, but the pumping efficiency is reduced because only a fraction of the radiation is useful for pumping at a given photon energy.

The signal to be measured is that of internal conversion electrons rather than the fluorescence of 8.4099-keV photons. There are two reasons for this. First, with a conversion ratio of 325, there are that many conversion electrons emitted for each fluorescence photon. Second, the fluorescence signal will not be able to compete with the scattered background radiation at the same photon energy emitted by the primary plasma. This is a major experimental problem because the plasma emission will be stronger than the nuclear fluorescence by many orders of magnitude. Thus, even the nuclear decay time's being ~4 times longer than the plasma emission time may prove insufficient to raise the fluorescence signal out of the background. The internal conversion electrons will be detected by a novel detector being developed for this experiment, in which thulium metal is deposited on the surfaces of a specially designed venetian-blind structure. The purpose of this design is to ensure that the short-range electrons can be extracted from the thin thulium layer and diverted by an electric field to a signal-amplifying channel-plate detector. The venetian-blind geometry prevents laser-target x rays from directly reaching the detector and thereby swamping the signal. Finally, to take advantage of the time difference between the nuclear decay half-life (3.9 ns) and the plasma emission time (~ 1 ns), a gating voltage will activate the detector several nanoseconds after the laser firing, further reducing the background.

The advantages of the thulium experiment can be summarized as follows:

- 100% isotopic abundance of ^{169}Tm, so no isotope separation is needed to increase pumping efficiency.
- Pumping energy is not too high (8.4099 keV).
- Lifetime (3.9 ns) longer than laser pulse width (0.6 ns).
- High signal of conversion electrons following x-ray pumping. For a 2.5-kJ target shot on OMEGA and a detector subtending 5% of the solid angle, 6×10^4 electrons will be detected.

The second experiment, involving the ^{137}La isotope of lanthanum, is similar in conception to that of the thulium experiment. Here too we pump a ground-state nucleus to a well-known excited state, at 10.6 keV. The required photon energy is somewhat higher than that required to excite ^{169}Tm. Furthermore, we have not as yet identified an intense x-ray line that can resonate with this transition, so we will have to rely on L-shell x-ray spectra of rare-earth ions that have densely spaced line spectra in this energy range. The predicted signal strength is higher than that from the thulium experiment by about a factor of 2, in spite of the weaker x-ray pump intensity. The conversion ratio (about 1) is also more favorable. Because of this and the very long half-life of the excited state (89 ns), a measurement of the 10.6-keV fluorescence should be easy. The main difficulty is the radioactivity of the ^{137}La, which has a half-life of 60,000 years. This makes the sample more costly than the thulium sample, and the mild radioactivity requires careful shielding. In both of these experiments, a single laser shot is sufficient to yield a discernible signal. However, mapping the decay and verifying the expected half-life may require several tens of laser shots.

The third experiment, excitation of isomeric ^{110}Ag (see Fig. 26.8), is much more costly and extensive and is more characteristic of future excitation experiments of nuclear candidates for γ-ray lasers. The first difficulty is the cost of obtaining the isomeric sample. Using a sample that was not subjected to isotope or isomer separation would be the least expensive. As explained, this will necessitate the accumulation of statistics over a large number of shots. The second difficulty is that the still incomplete knowledge of the energy-level scheme renders uncertain whether a close level permits realistic pumping from the isomeric state. The 3+ level is connected by a highly forbidden transition to the 6+ isomeric state. Extrapolation of known energy-level schemes lends credence to speculation that a level with spin closer to six may lie within about 10 keV or less to the isomeric state. Covering the x-ray energy range up to about 10 keV will require several laser shots using up to ten different targets. If excitation occurs, detecting the resulting fluorescence will be relatively easy. First, the fluorescence occurs at a vastly higher photon energy than that of the x-ray pump, reducing background problems. Second, ^{110}Ag in the ground state is unstable and decays with a convenient, 24.4-s lifetime into ^{110}Cd, providing an additional measurement of the β-decay electrons.

The successful outcome of this and similar experiments, in effect, constitutes experiments in nuclear spectroscopy: the energy and half-life of previously inaccessible levels are being determined. Such data can be used in designing optimal conditions for γ-ray lasing.

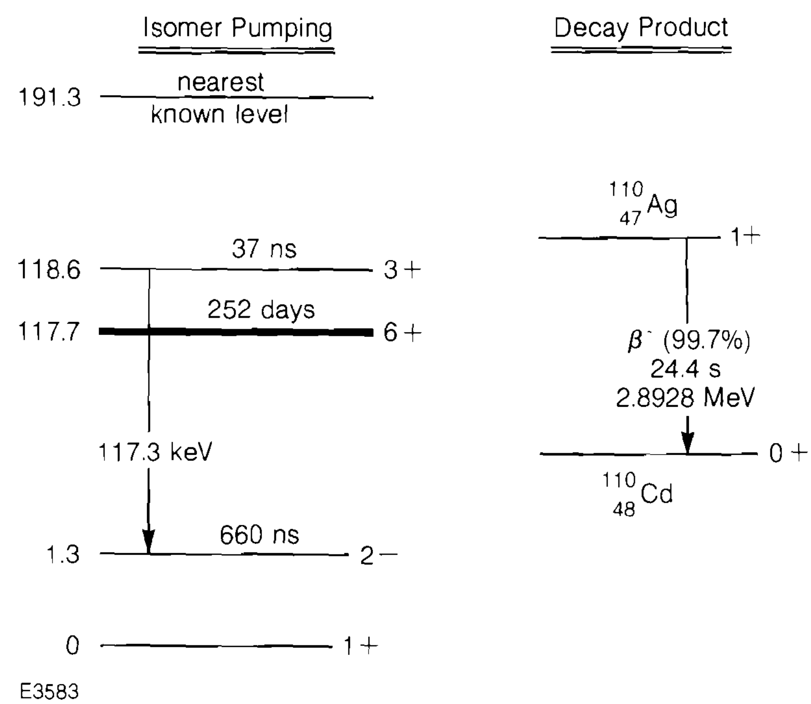

Fig. 26.8
Nuclear energy levels in ^{110}Ag. Excitation of the isomer of 252-days half-life is an example of promising nuclear pumping schemes. Once in the 1+ state, the nucleus decays via β-decay into $_{48}Cd$.

OMEGA Experiments

Experiments have begun at LLE to analyze the suitability of the x-ray pulse emitted from high-power laser-irradiated targets for exciting nuclear transitions. These experiments employed the 24-beam OMEGA laser system. For irradiating a nuclear sample with x rays from an adjacent target, 12 beams can be focused onto a flat target or a spherical target of large radius. The experiments described here were performed mostly in the former mode. In future experiments, an elongated lasing medium, where x-ray gain measurements would be attempted, is required. In this case, a multidirectional irradiation of the x-ray source in a cylindrical geometry is necessary. Such a capability has, in fact, been developed[4] on OMEGA for x-ray laser experiments.

The recent conversion of all 24 OMEGA beams into the UV (λ = 351 nm), by frequency tripling, greatly enhances x-ray production efficiency[5] and, therefore, OMEGA's usefulness for nuclear excitation experiments. In addition to high x-ray fluence, such experiments require a wide x-ray spectral coverage to maximize the chance of resonantly exciting nuclear levels. This is essential because the location of nuclear levels is not generally well known. Thus, a continuous x-ray spectrum extending up to 10 keV could locate nuclear levels that can be pumped in a 10-keV interval extending above a ground (or an isometric) level.

The spectrum emitted by a laser-irradiated target consists of both a continuum and spectral lines. Whereas isolated spectral lines can be useful only if an occasional resonance with a nuclear transition occurs, densely spaced lines from appropriately chosen targets can serve as a quasi continuum. This adds another reason for frequency tripling, since irradiation by 351-nm light reduces preheating by long mean-free-path electrons. These electrons interact with cold material and give rise to a narrow K_α line, where the chances for coincidence with nuclear transitions are very small. On the other hand, a short-wavelength laser is coupled into a hot plasma, where the lines are broad; and a multiline spectrum, which can become a quasi-continuum, is emitted. Additionally, the virtual absence of fast electrons greatly reduces background at the energy of typical γ-fluorescence emission.

The x-ray emission from a UV-laser-irradiated target is mostly the thermal emission from a plasma in the 1-keV to 3-keV temperature range. This emission ranges up to about 10 keV in photon energy, and a laser plasma's usefulness as a source declines for much higher photon energies. On the other hand, as shown below, x-ray fluence levels as high as 10^{16} keV/keV to 10^{17} keV/keV can be achieved with a UV laser of only 1-kJ energy, over selected energy intervals below 10 keV. By choosing several target materials appropriately, one can completely cover the x-ray energy range out to 10 keV or somewhat higher energies.

The x-ray continuum (due to free-bound and free-free transitions) generally falls off with increasing photon energy, except at ionization edges. It is strongest within the sub-keV energy range because this part of the spectrum is emitted by the colder, denser target layers of the interaction region. At photon energies below about 0.5 keV, the emission is that of a blackbody. This can be seen from the formula[6] for opacity τ (absorption coefficient times length ΔR) at the ionization edge of a level of quantum number n in a hydrogenic ion of charge Z:

$$\tau = \frac{8}{3^{3/2}\,\pi^2}\;\frac{h^3\,g\,n}{m^2\,c\,e^2\,Z^2\,M_i}\;\rho\Delta R \quad . \tag{2}$$

Here, g is the Kramers-Gaunt factor, M_i is the ionic mass, ρ is the mass density, and all physical constants have their usual designations. It can be easily shown that Eq. (2) can be approximated by

$$\tau \sim 10^3\,\rho\Delta R/(TE^{1/2}) \quad , \tag{3}$$

where the relevant photon energy E and the temperature T are expressed in keV. For blackbody conditions to hold, τ should be larger than 1. For the interaction region in the target, $\rho\Delta R$ is of the order of 10^{-4} – 10^{-3} g/cm^2 and $T \leq 1$ keV, so that blackbody radiation (which peaks at $E_m \sim 3T$) occurs for photon energies less than about

0.3 keV to 0.7 keV. The radiation intensity of a blackbody can be written as

$$I \sim 10^{17}\, T^4 \ \mathrm{W/cm^2} \quad , \tag{4}$$

where T is in keV. Thus, for T = 0.2 keV, the blackbody intensity (10^{14} W/cm^2) is a sizable fraction of the incident laser irradiance (typically $10^{14} - 10^{15}$ W/cm^2). High conversion efficiency (up to 50%) of the incident short-wavelength laser into sub-keV x rays has been extensively documented in the literature.[7] These high efficiencies translate to a spectral intensity in the sub-keV region of up to 10^{18} keV/keV, using a 1-kJ, short-wavelength laser. For higher photon energies, the spectral intensity as described below is considerably lower, but still substantial.

We now show results of spectra obtained on OMEGA, using either a flat-crystal x-ray spectrograph (Fig. 26.9) or a Von Hamos-type[8] focusing crystal spectrograph (Figs. 26.10–26.12). The x-ray fluence was calculated using measured crystal integrated reflectivities and published film calibration employing continuous wave x-ray sources. The uncertainty in the results due to both of these calibrations is estimated to be less than a factor of 2.

The compression of spherical targets can result in an intense x-ray continuum due to the higher density achieved at the compressed target core.[9] We show in Fig. 26.9 spectra obtained by imploding glass shell targets filled with a deuterium-tritium mixture, with and without argon, using the OMEGA laser system without the frequency tripling (λ = 1054 nm). For UV-laser irradiation (the remaining data shown here), the density in the interaction region is itself quite high ($\geq 10^{22}$/cm^3), leading to high x-ray conversion efficiency even with no target implosion. However, UV-laser-driven implosion has the potential for still higher x-ray yield than that found in the experiments described below. The strong and smooth continuum is very useful for attempting the excitation of nuclear levels of poorly known location. Evidently, the continuum extends to energies above 3.5 keV; the cutoff around that energy is due to the edge of the diffracting crystal.

The continuum radiation in Fig. 26.9 is predominantly due to free-bound (recombination) transitions into Si^{+14} ions, emitted by the imploded part of the glass shell. Extending such continua to higher photon energies requires targets with higher-Z materials. This is limited by the temperature achievable at a given laser power, which has to be high enough to cause significant ionization of these higher-Z species. Experiments indicate that recombination continua of higher-Z elements up to photon energies of about 5 keV can contain significant intensity.

To extend the useful emitted x-ray spectrum to much higher photon energies, one must rely on line spectra. In general, by progressing to targets of higher-Z ions, the line spectrum changes from K- to L-, M-, and finally N-shell transitions. Here K-shell spectra mean transitions from the L-shell (n = 2) and higher shells to the K-shell, where the L-shell is mostly empty. L-shell spectra mean transitions from the M-shell and higher shells to the L-shell, where the K-shell is fully occupied and the

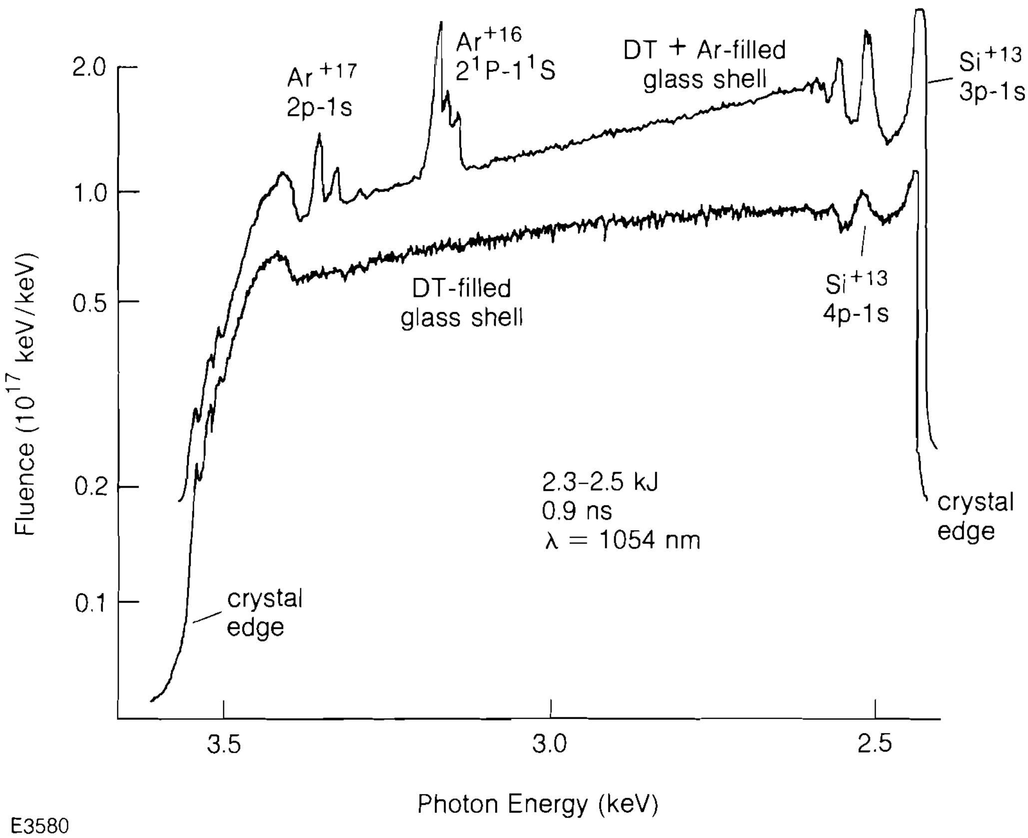

Fig. 26.9
Continuous x-ray spectra emitted from two target implosions on OMEGA. Twenty-four beams at λ = 1054 nm were used. The strong and smooth continuum is useful for attempting the excitation of nuclear levels of poorly known location.

M-shell mostly empty; thus, as one moves from K-shell to higher-shell spectra, the density of lines increases but their individual intensity, in general, declines. Therefore, for maximizing the likelihood of hitting an unknown nuclear resonance, a higher-Z target is preferable. However, a strong K-shell line is preferable when it is known to resonate with a given nuclear transition.

A case of K-shell line resonance exists between the 1^1S–2^1P line of Cu^{+27} at 8.394 keV and the transition in ^{169}Tm between the +1/2 ground level and the +3/2 level at 8.4099 keV above the ground level. This excitation is the goal of an experiment currently under way, using the OMEGA laser system to generate x rays. The line spectra in the 1-keV to 10-keV range from laser-irradiated targets of low to medium Z ($Z \lesssim 30$) are dominated by the helium-like species (e.g., Cu^{+27}), of which the 1^1S–2^1P is the strongest line.

In Fig. 26.10 we show the spectrum emitted from a copper-coated CH ball target, irradiated by the 24 OMEGA beams. The target diameter was 582 μm, overcoated with a 1-μm thickness of copper. The laser pulse, 1.46 kJ in 0.6 ns, irradiated the target symmetrically over the full solid angle, so that no more than half of the laser energy irradiated the side of the target facing the spectrometer. Each beam was focused to a focal spot of ~50-μm diameter on the surface of the target, making the irradiance about 5×10^{15} W/cm^2. The width of the lines in these spectra is determined by the source size (since neither spectrometer focuses the radiation in the dispersion direction). The true spectral width of an individual line in Fig. 26.10, about 7 eV, is due mostly to Doppler broadening. However, the feature marked as the $1s^2$–$1s2p$ transition of Cu^{+27} actually includes the resonance line 1^1S–2^1P, the intercombination line 1^1S–2^3P, as well as several dielectronic satellites in Cu^{+26}. A high-resolution recording of a similar spectrum (see Ref. 8b, Fig. 9) shows that a group of several lines of comparable intensity occupy an interval of about 80 eV. High resolution is achieved by irradiating a small target or using only one laser beam which is focused on the surface of the target. This is a large fraction of the width of the spectral feature at 8.4 keV, which would be ~120 eV if it corresponded entirely to spectral broadening. Part of the sloping continuum is caused by fluorescence within the spectrograph and does not represent target emission. The fluence of 1.2×10^{17} keV/keV marked for the resonance line is obtained by dividing the net intensity in the observed spectral feature by the combined estimated Doppler widths of the components comprising this feature. The uncertainty in this figure is smaller than a factor of 2.

Fig. 26.10
X-ray spectrum from a copper-coated CH ball target, irradiated by 24 OMEGA beams at λ = 351 nm. The Cu^{+27} line at 8.394 keV resonates with a transition from the 1/2 + ground nuclear level of ^{169}Tm to the 3/2 + excited state at 8.4099 keV (with Doppler shift). The x-ray fluence value for the resonance line is into 4π solid angle.

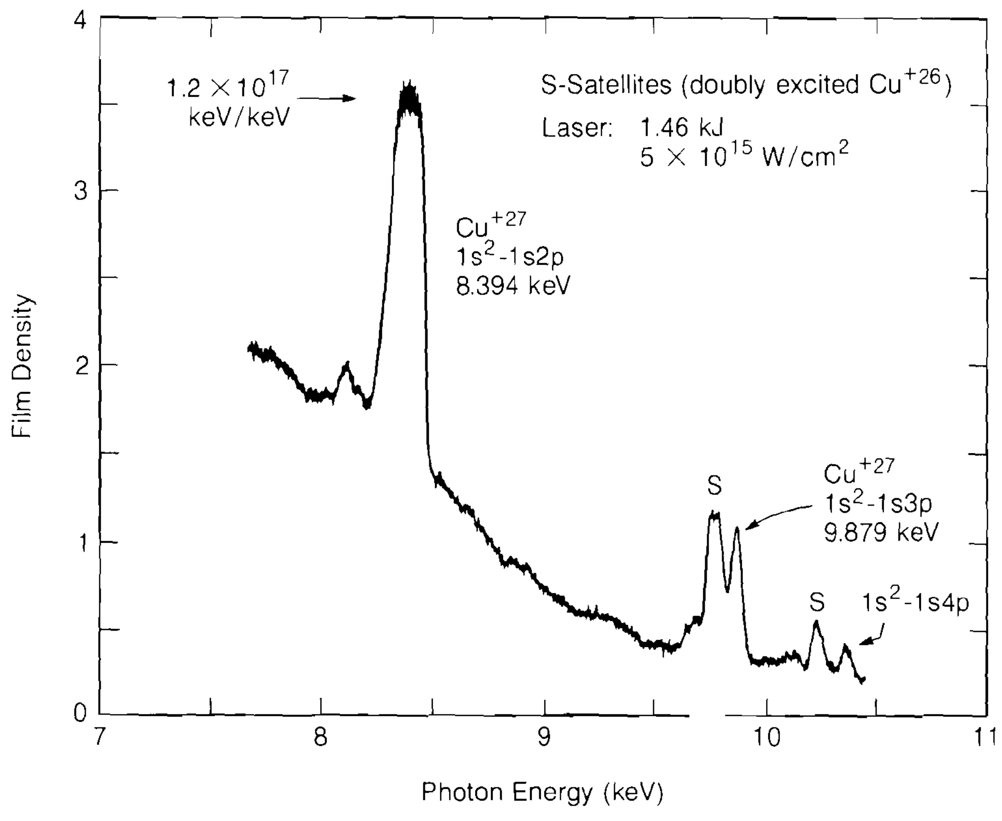

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Figure 26.11 shows the spectrum obtained from a target similar to that of Fig. 26.10, after the laser energy was raised to 1.83 kJ. The resonance line here is sufficiently intense to cause film saturation (and therefore broadening). The fluence on this line is higher than 2×10^{17} keV/keV. Now not only the resonance line, but also higher energy lines, become sufficiently intense for nuclear pumping experiments.

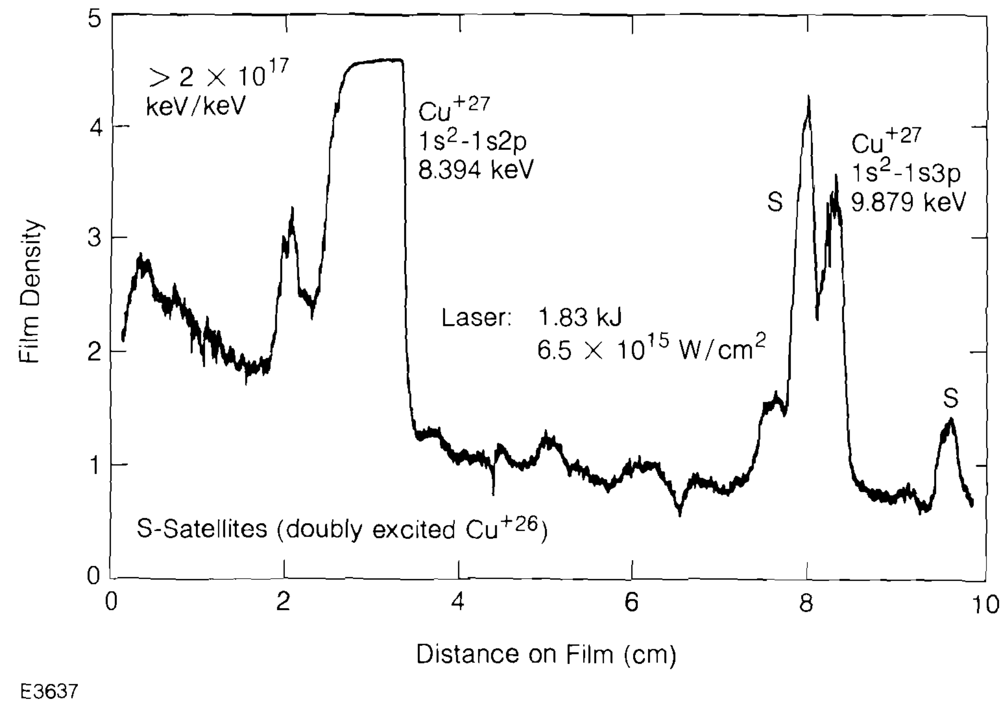

Fig. 26.11
X-ray spectrum from a similar experiment to that of Fig. 26.10, where the laser energy was raised to 1.83 kJ. The fluence within the Doppler width of the resonance line of Cu^{+27} is higher than 2×10^{17} keV/keV into 4π solid angle.

To increase the spectral coverage in the spectral range above about 5 keV (where continua are not very intense), one needs to go from K-spectra (like those of Figs. 26.10 and 26.11) to L-spectra. We have chosen a target material, praseodymium (Z = 59), whose L-spectrum falls around the 8.4-keV energy. The ionization energies of praseodymium ions with outer electrons in the L-shell range from 8.8 keV to 11.6 keV. The lines due to transitions from higher shells into the L-shell would fall roughly within the 5-keV to 11-keV range. The measured range of intense line radiation (Fig. 26.12) is about 7.6 keV to 8.6 keV. The low-energy limit is simply determined here by the edge of the diffracting crystal; the actual spectrum undoubtedly extends to lower energies. The high-energy limit is determined by the insufficiency of the plasma temperature to cause higher-energy excitations. The irradiation configuration in this experiment comprised 12 OMEGA beams; the target was moved away from the incoming beams (and the spectrograph) so that the beams converged to one focal spot of 180-μm diameter, yielding an irradiance of 6×10^{15} W/cm^2. Focusing to a smaller spot may result in a higher temperature and the extension to higher photon energies of the spectrum in Fig. 26.12. For the present

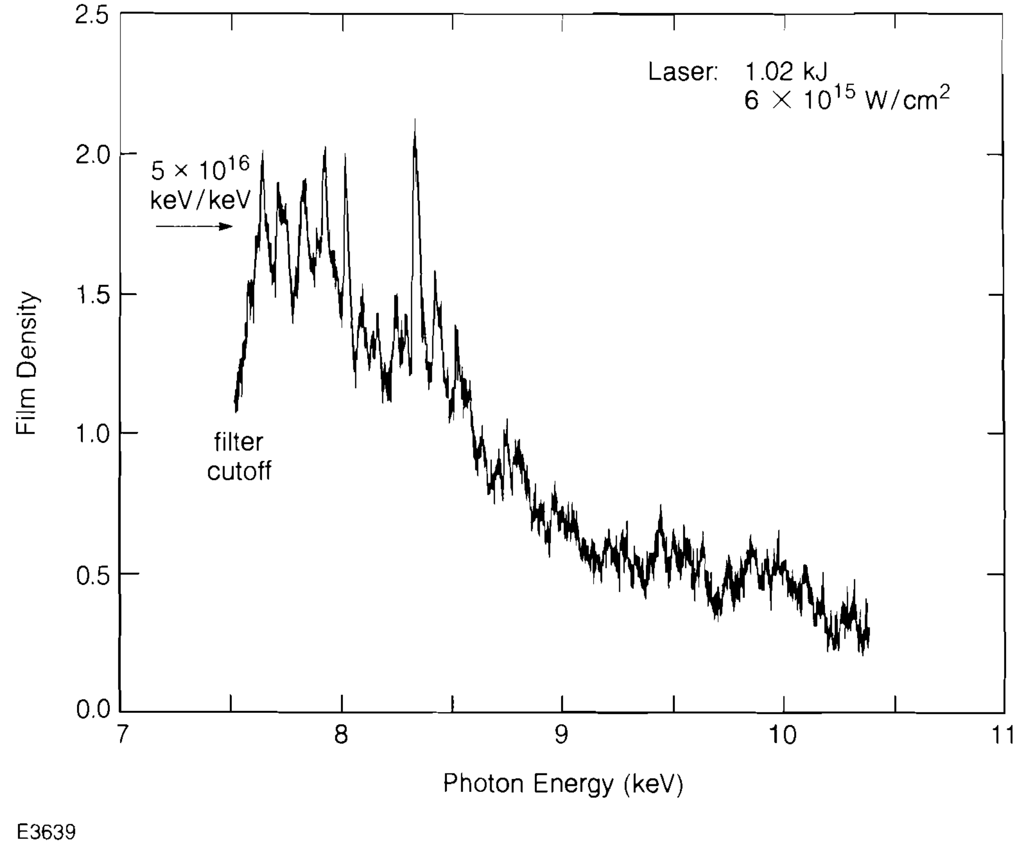

Fig. 26.12
L-shell spectrum from a praseodymium target irradiated by 12 OMEGA beams at λ = 351 nm. The 12 beams were all focused to the same focal spot of diameter 180 μm on the surface of the target.

experimental conditions we can estimate the energy interval containing the group of intense lines as extending from 6 keV to 8.5 keV. The estimated fluence of the strong lines in Fig. 26.12, 5 $\times$ 10^{16} keV/keV, is still substantial enough for significant nuclear excitation. The net coverage of the above-stated energy interval is only about 20%. This estimate is based on the true spectral widths (due mostly to the Doppler effect), which were also used to deduce the fluence estimate. If we performed experiments with rare-earth targets of successively higher Z, the spectral lines of a given type would move up in energy by about 0.3 keV for a one-step progression in Z. If the energy range covered by each target is about the same as that estimated for praseodymium (namely, 2.5 keV), a given interval in the spectrum will include spectral lines from eight successive rare-earth targets. Since the line spectrum from a single target covers about 20% of the energy range over which it extends, the cumulative effect of eight successive targets will completely cover that range. Complete coverage with fluence higher than 10^{16} keV/keV, using this procedure, is expected to apply to at least the 5-keV to 10-keV energy range. Higher energies will be accessible if higher temperatures than those achieved here can be achieved, e.g., by tighter focusing.

In conclusion, experiments on the OMEGA laser system can produce x-ray fluences of at least 10^{16} keV/keV, or about 1 J/keV, in one shot, for photon energies of up to about 10 keV. X-ray fluences of this order of magnitude are surmised to be sufficient to induce nuclear excitation in several nuclei, either in the ground state or in an isomeric state, to an extent permitting reliable measurement of the resulting nuclear fluorescence (gamma photons or internal conversion electrons). However, the radiation has to cover an energy range that includes the energy of the nuclear transition to be pumped. We showed that in the range up to about 10 keV OMEGA can achieve this condition with either one target or a sequence of up to ten different targets. Further work on optimizing the production of x rays by changing the laser and target parameters could possibly increase the x-ray fluence and extend it to above 10 keV.

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