frequency changes during the electronic transition) as well as medium anharmonicity, increase the mean effective phonon frequency (due to the appearance of higher harmonics) and, therefore, are expected to moderate the calculated energy-gap dependence of the rate.[8]

(d) In principle slow processes can be masked by the occurrence of competing processes like infrared emission, pure vibrational relaxation, and energy transfer to other impurities. In the present case pure intrastate (infrared radiation or vibrational relaxation) relaxation was shown to be too slow to compete with the observed rates. The lack of concentration dependence of the rates also excludes energy transfer between different CN molecules.

In summary, we have established the importance of a process characterized by interstate cascading for the relaxation of diatomic guest molecules. The observed rates in CN qualitatively satisfy the energy-gap law but further studies are needed to establish quantitative agreement.

One of us (A.N.) is grateful to Professor J. Jortner for many helpful discussions.

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First Operation of a Free-Electron Laser*

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A free-electron laser oscillator has been operated above threshold at a wavelength of 3.4 μm.

Ever since the first maser experiment in 1954, physicists have sought to develop a broadly tunable source of coherent radiation. Several ingenious techniques have been developed, of which the best example is the dye laser. Most of these devices have relied upon an atomic or a molecular active medium, and the wavelength and tuning range has therefore been limited by the details of atomic structure.

Several authors have realized that the constraints associated with atomic structure would not apply to a laser based on stimulated radiation by free electrons.[1-5] Our research has focused on the interaction between radiation and an electron beam in a spatially periodic transverse magnetic field. Of the schemes which have been proposed, this approach appears the best suited to the generation of coherent radiation in the infrared, the visible, and the ultraviolet, and also has the potential for yielding very high average power. We have previously described the results of a measurement of the gain at 10.6 μm.[6] In this Letter we report the first operation of a free-electron laser oscillator.

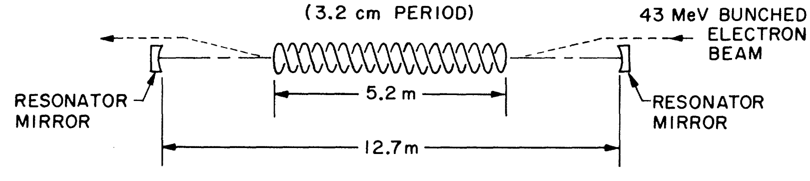

FIG. 1. Schematic diagram of the free-electron laser oscillator. (For more details see Ref. 6.)

TABLE I. Laser characteristics.

Laser characteristics	Above threshold	Below threshold
Wavelength (μm)	3.417	3.407
Width (full width of half-maximum)	0.008	0.031
Average Power (W)	0.36	10^{-8}
Peak power (Ref. 7)	7×10^{3}	10^{-4}
Mirror transmission	1.5%	

Our apparatus is shown schematically in Fig. 1. A superconducting helix generates a periodic transverse magnetic field of 2.4 kG. A 43-MeV electron beam from the superconducting accelerator is fired along the axis of the helix. Radiation passing through the helix with the electron beam is amplified and a pair of mirrors at the ends of the interaction region provide feedback.

The characteristics of the oscillator are listed in Tables I and II. The wavelength was 3.417 μm and the average power output was 0.36 W. Factoring out the duty cycle of the machine,[7] this translates to a peak power of the order of 7 kW. With a mirror transmission of 1.5% the intracavity power was 500 kW. The total energy collected on the detector was 0.01% of the electron beam energy.

The laser spectrum is shown in the upper half of Fig. 2 and the spontaneous spectrum in the lower half. Note the difference in the radiated power: Above threshold, the oscillator power increases by a factor of 10^8 over the spontaneous radiation. The oscillator linewidth was 8 nm (200 GHz).

The electron energy in the experiment was chosen to satisfy the wavelength equation[6]

$$\lambda = \frac{\lambda_q}{2\gamma^2}\left[1+\frac{1}{4\pi^2}\left(\frac{\lambda_q^2 r_0 B^2}{mc^2}\right)\right],$$

where λ_q is the magnet period, γmc^2 the electron energy, r_0 the classical electron radius, and B the magnetic field strength. The wavelength varies inversely as the square of the electron energy.

The experiment demonstrates the capability of a free-electron laser to operate at high power at an arbitrary wavelength. We note that the linewidth observed in the experiment is by no means the limiting linewidth. As established by the earlier experiments,[6] a homogeneously broadened gain profile is attainable and the laser linewidth can be improved by means of intracavity dispersive elements. The efficiency of the present laser is limited by the fraction of the electrons' energy which can be converted to radiation in a single pass through the interaction region. This limitation would not apply to devices in which the electron beam was reaccelerated and recirculated through the interaction region as in an electron storage ring, where efficiency above 20% should be possible.[9] The nanosecond electron bunch

TABLE II. Electron beam characteristics.

Energy (Ref. 8)	43.5 MeV
Width (full width at half-maximum):	0.05%
Average current	130 μA
Peak current (Ref. 7)	2.6 A
Emittance (at 43.5 MeV):	0.06 mm mrad

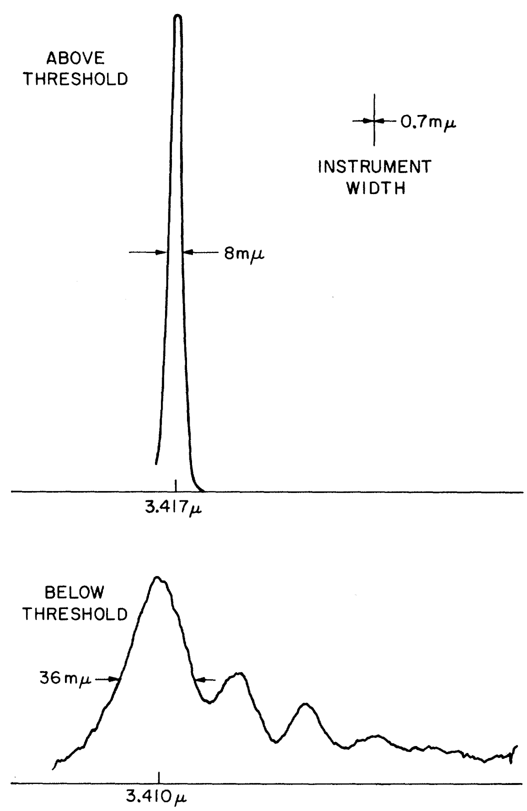

FIG. 2. Emission spectrum of the laser oscillator above threshold (top) and of the spontaneous radiation emitted by the electron beam (bottom).

lengths attainable in storage rings would further improve both the linewidth and the average power output.

Because the gain falls at short wavelengths, a higher electron current will be required to support laser operation in the visible and the ultraviolet. Based on the small-signal gain equations,[4,10,11] sufficient current has been stored in existing electron storage rings to sustain laser operation at wavelengths as short as 1200 Å.[9]

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Optical Second-Harmonic Generation in Gases: "Rotation" of Quadrupole Moment in Magnetic Field

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We report optical second-harmonic generation in gases in a transverse dc magnetic field. An electric quadrupole moment of sodium or calcium vapor is induced by a two-photon transition. The effect of the magnetic field is to transform the wave function within both the ground and the excited magnetic sublevels, and to make a new component of the quadrupole moment which can generate a collinear second harmonic. Transverse magnetization is not important in the present case.

Optical second-harmonic generation (SHG) in centrosymmetric media is possible only under certain restricted conditions. The experiments so far reported can be grouped by the conditions employed: (1) The second harmonics from the electric quadrupole moment induced by noncollinear fundamental fields were observed in an anisotropic crystal[1,2]; (2) those from the electric quadrupole and the magnetic dipole interaction were observed on the boundary surfaces of isotropic materials[3,4]; and (3) SHG by a dipole interaction at a noncentrosymmetric surface layer was deduced from an experiment on an adsorbed surface layer.[5] Bethune, Smith, and Shen[6] recently demonstrated sum-frequency generation with a resonant noncollinear excitation in sodium vapor. SHG was not possible in the $3S$-$4D$ transition. Hänsch and Toschek,[7] on the other hand, discussed a collinear three-wave mixing in the background medium with transverse magnetization.

In this Letter we describe an experiment in