Recent, dynamic nuclear polarization enhanced nuclear magnetic resonance (DNP/NMR) has emerged as a powerful technique to obtain significant enhancements in spin spectra from biological samples. For DNP in modern NMR systems, a high power continuous-wave source in the submillimeter wavelength range is necessary. Gyrotrons can deliver tens of watts of CW power at submillimeter wavelengths and are well suited for use in DNP/NMR spectrometers. To date, 140 GHz and 250 GHz gyrotrons are being employed in DNP spectrometer experiments at 200 MHz and 380 MHz at MIT. A 460 GHz gyrotron, which has operated with 8 W of CW output power, will soon be installed in a 700 MHz NMR spectrometer. High power radiation with good spectral and spatial resolution from these gyrotrons should provide NMR spectrometers with high signal enhancement through DNP. Also, these tubes operating at submillimeter wavelengths should have important applications in research in physics, chemistry, biology, materials science and medicine.

Keywords: Gyrotrons, Continuous-wave (CW), Submillimeter-wave, Dynamic Nuclear Polarization (DNP), Nuclear Magnetic Resonance (NMR)

1. INTRODUCTION

Radiation sources with good spectral characteristics and reasonable output power at submillimeter wavelengths are needed for a great number of researches and practical applications [1]. However, coherent sources of high quality radiation in this area are scarce. For radiation sources based on optics, power decreases with decreasing frequency toward the THz region. For radiation sources based on microwave solid-state electronics, power decreases with increasing frequency toward the THz region [2]. Therefore, a considerable number of studies have been conducted to develop radiation sources in the THz frequency region [1]. Nevertheless, most of the currently available sources still do not satisfy the requirements for recent applications, such as dynamic nuclear polarization enhanced nuclear magnetic resonance (DNP/NMR) [3, 4] and electron paramagnetic resonance (EPR).

DNP is an electron-nuclear resonance technique that can significantly enhance the signal-to-noise ratio in NMR experiments. By transferring the highly populated spin polarization of the electron to the less populated nuclear one, the acquisition time in NMR experiments is reduced, which broadens the applicability of NMR in bio-molecular systems. To advance this technique by increasing the spectral resolution, high magnetic field strength is required. However, the corresponding electron spin resonance frequency typically falls in the submillimeter wavelength range. Therefore, DNP/NMR experiments require continuous-wave submillimeter wave sources with very high stability in the phase, frequency, and amplitude. Consequently, there has been a renewal of interest in the cyclotron-resonance maser (gyrotron) that has been developed for plasma physics applications. Indeed, gyrotrons have reached frequencies up to 0.89 THz [5]. Also, they can deliver tens of watts of CW power at submillimeter wavelengths and are well suited for use in DNP/NMR spectrometers.

The gyrotron is a vacuum electron device employing a strong magnetic field, an electron beam, and a resonant cavity, as shown in Fig. 1. The electrons emitted from the cathode acquire a transverse and axial velocity due to the electric and magnetic fields in the gun region. Then the electrons move toward the cavity in the growing magnetic field, where the electron flow undergoes the adiabatic compression and the electron orbital momentum increases. In the region of...
uniform magnetic field, the electron's orbits are perturbed at the Doppler shifted cyclotron frequency (or its harmonic) which is also resonant with an eigen-mode supported by the cavity. Along the cavity, the change of the relativistic mass of the electrons by the perturbation leads to the instability for powerful self-oscillation, which transforms a part of the kinetic energy of the electrons into the microwave energy [6]. After the interaction, the generated electromagnetic wave is separated from the electron beam by a mode converter and extracted through the output window. Then the spent beam is decompressed in the decreasing magnetic field and collected by the beam collector.

Using a smooth wall interaction structure, whose transverse dimensions are many wavelengths of the operating frequency, gyrotrons have the ability to generate very high power when compared to slow wave microwave tubes such as klystrons, backward wave oscillators or solid state devices. Operating at higher voltages (typically above 10 keV) than the energy level of the radiated electromagnetic wave (~ meV), the weakly non-equidistant energy spectrum of electron cyclotron radiation allows initially populated levels and, correspondingly, high efficiency of gyrotrons (electron cyclotron maser) contrary to lasers in the submillimeter regime [6]. In spite of these capabilities, only a small number of research programs have been devoted to gyrotrons for submillimeter wave applications.

In this article, we review the present status of compact CW gyrotrons developed for DNP/NMR spectroscopy at MIT, and discuss the spatial, operational and spectral characteristics of the gyrotrons for possible applications, such as medical imaging and diagnostics.

Figure 1 Schematic drawing of a gyrotron in cross-sectional view (not shown to scale). The basic components of the gyrotron include a magnet, and an electron gun, and a vacuum tube which consists of a beam tunnel, a resonator, and a mode converter and collector.

II. MIT GYROTRONS FOR DNP/NMR

To date, 140 GHz and 250 GHz gyrotrons are being employed in DNP experiments at 200 MHz and 380 MHz spectrometers at MIT [7-9]. Also, a 460 GHz CW gyrotron oscillator operating at the second electron cyclotron
The 140 GHz gyrotron is capable of producing 15 s long output pulses at 50% duty cycle. For single pulse operation, an output pulses as long as 5 min is easy to achieve. The output in the TE_{03} mode is converted into the TE_{01} mode by a mode converter. Outside of the gyrotron, the TE_{01} mode is converted to the TE_{11} mode using a serpentine rippled wall mode converter [7].

The 250 GHz gyrotron and the 460 GHz gyrotron are true CW sources of which operation lasting days is possible under computer control. They are equipped with similar magnets which have cross-bores for effective separation of the generated electromagnetic wave and electron beams. The difference is the harmonic number of the electron cyclotron frequency. The 460 GHz gyrotron adopts the second harmonic to mitigate the limiting factor of superconducting magnet technology to achieve a high field for fundamental mode operation at this frequency. The outputs of the 250 and the 460 GHz gyrotrons are converted into the HE_{11} mode (Gaussian beam) by the Vlasov type mode converters [6].

**Table 1** Operational parameters and resulting output powers and efficiencies of MIT gyrotrons for DNP/NMR experiments.

<table>
<thead>
<tr>
<th></th>
<th>140 GHz</th>
<th>250 GHz</th>
<th>460 GHz</th>
</tr>
</thead>
<tbody>
<tr>
<td>Operation voltage, Vo (kV)</td>
<td>12</td>
<td>12</td>
<td>12</td>
</tr>
<tr>
<td>Beam current, Io (mA)</td>
<td>25</td>
<td>25</td>
<td>135</td>
</tr>
<tr>
<td>Operating mode</td>
<td>TE031</td>
<td>TE032</td>
<td>TE061</td>
</tr>
<tr>
<td>Tube output mode</td>
<td>TE01</td>
<td>HE11</td>
<td>HE11</td>
</tr>
<tr>
<td>Magnetic field, Bo (T)</td>
<td>5.1</td>
<td>9.0</td>
<td>8.4</td>
</tr>
<tr>
<td>Cyclotron harmonic number</td>
<td>1</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>Output power (W)</td>
<td>14</td>
<td>7</td>
<td>8</td>
</tr>
</tbody>
</table>

**III. SPATIAL CHARACTERISTIC**

![Image of radiation intensity pattern](image)

Figure 2 Radiation intensity pattern from the 460 GHz gyrotron; (a) burn image on thermal paper with absorbing material on the back side and (b) image recorded by a pyroelectric camera.

The output intensity patterns were measured using three techniques; thermal paper, liquid crystal paper, and an array of pyroelectric sensors (the Pyrocam III pyroelectric camera detector from Spiricon, Inc. [12, 13]). Figure 2 shows the 460 GHz gyrotron mode pattern at the end of a 1.6 m long, 19 mm diameter corrugated waveguide which is matched to the gyrotron window. The pattern on thermal paper with absorbing material on the backside (a) has several peaks. Focusing on the strongest peak with the pyroelectric array camera, the image (b) shows a Gaussian-like pattern. This camera consists of an array of 124-by-124 LiTaO3 pyroelectric sensors with spacing between each pixel of 100 μm. There is a motorized chopper blade over the sensor array to modulate the beam for detection of small CW signals (2.2 mW/cm²). Similar mode pattern images have also been made with both the 140 GHz and the 250 GHz gyrotrons [7, 8].
The measured pattern matches reasonably well with the theoretical output mode. The output pattern from the 250 GHz gyrotron shows a small sidelobe and the measured beam is slightly elliptical. These results indicate that gyrotrons have reasonably good spatial characteristics enough to couple the output power to the transmission system maximizing power concentration or spatial resolution in practical applications.

IV. CW STABILITY

DNP requires continuous strong submillimeter wave irradiation to pump up the unpaired electron spin state under a strong magnetic field. Therefore, continuous operation of gyrotrons for a day long period (or longer) is required with very high stability in the phase, frequency, and amplitude. We report extension of the stable operation of the 460 GHz gyrotron for a run of 24 hours, thus allowing its application to DNP/NMR experiments.

![Graphs showing CW stability](image-url)

**Figure 3** Summary of stable CW operation of the 460 GHz gyrotron over 24 hours: (a) Diode voltage proportional to the output power. (b) Pressure on the ion pumps located at the collector side. (c) Water-cooled collector temperature. (d) Cathode heater current. (e) Collector current. (f) Body current.

For a gyrotron the output power is a direct function of the beam current, which suggests that feedback control of the beam current should be sufficient to regulate the output power. The gyrotron is controlled by a LabVIEW-based control system on a PC. The control system has separate software and hardware safety interlocks monitoring the coolant flow rates and temperature, the pressure in the tube, and the protection for the magnetron injection gun. To achieve a low base-pressure which is necessary for multiple hour CW operation, we baked the tube at 120 °C for about 80 hours, and processed by slowly increasing the beam current at low voltages below 100 V for about 40 hours. After seeing that the pressure and the coolant temperatures were stable with more than 4 W of CW output power at about 1.7 kW collector load, we attempted the 24-hour completely computer controlled run (Fig. 3). The power was held constant at 3.1 W by feedback regulation, i.e., proportional, integral, and derivative (PID) adjustments to the filament current. The output power was monitored by a diode detector and referenced at the beginning and end of the monitoring period with a dry calorimeter.

From the histogram analysis for the diode voltage fluctuation over 24 hours, it is shown that the power was maintained stable and the power fluctuations are normally distributed within ±0.5%, where the sampling period was 900 ms. Similar measurements have been done for the 250 GHz gyrotron for the period of up to 10 days, and the...
output power was stable within 0.8%. Therefore, the stability requirement for DNP/NMR experiments is able to be achieved by the computer controlled operation of gyrotrons. It is clear that this is a promising characteristic of gyrotrons for extended applications, such as imaging systems for security and medical therapy.

Figure 4 Statistical analysis of power fluctuations from the 460 GHz gyrotron over one day long operation.

V. SPECTRAL CHARACTERISTICS

The DNP enhancement factor is a function of the gyrotron frequency. Consequently, the frequency should be stable within the allowed bandwidth, which is determined by the spin resonance spectrum of the paramagnetic material providing unpaired electrons for DNP. In addition, as frequency goes up, that is, the NMR magnetic field increases, the relaxation time of the spin system becomes long. Therefore, the resonance condition becomes much narrower, and typically frequency stability of less than 1 MHz is required for appropriate NMR signal enhancement.

To see the spectral characteristics of the gyrotron, we used the 140 GHz gyrotron operating in pulses with a duration of 30 s. A harmonic heterodyne receiver system was used, in which the receiver employs a low frequency local oscillator (LO) of between 8-26.5 GHz and a harmonic mixer to mix the gyrotron signal with the 7th harmonic of the LO (Micro-Lambda YIG oscillator). The LO frequency is counted by a frequency counter (Phase Matrix EIP 578 B) and locked by a phased locked loop stabilizing the LO to 10 kHz. The intermediate frequency (IF) from the mixer is amplified by a series of low noise amplifiers and detected by a spectrum analyzer (SA) with the dynamic range of 90 dB. To keep the 10 kHz resolution of the locked LO, the sweep time of the SA is set to above 1 ms and the resolution bandwidth is fixed at 10 kHz. The gyrotron output is sampled by a mirror and a 1.5 meter long Ka-band waveguide and delivered to the area where the fringe magnetic field intensity is below 5 gauss.

Figure 5 shows the frequency variation over the duration of the gyrotron pulse. Because a considerable portion of the generated radiation power is dissipated by heating the cavity wall, which results in the thermal expansion of the cavity, there is a down-shift of frequency. A transient frequency shift of 12 MHz has been observed. In comparison with the exponential fitting curve, the time constant is estimated to be approximately 3 s. After 10 s, the frequency is saturated and stabilized within 1 ppm. The sensitivity of the frequency shift due to thermal expansion is measured to be -2 MHz/K by changing the temperature of the cavity coolant. In cooperation with a re-circulating chiller with the
temperature precision of ±0.1 degree, the expansion of the cavity during operation reaches equilibrium, where the radiation frequency is maintained constant. Indeed, during the days-long CW operation of the 250 GHz gyrotron and 460 GHz gyrotron, no significant changes in the frequency were observed except for initial transient periods.

Figure 5 Frequency shift over the duration of the pulse in the 140 GHz gyrotron operation. Solid dots represent measured data and the solid line is a curve fitted to an exponential function. For this measurement, the gyrotron operates at 12.9 kV of voltage and 30 mA of beam current. The spectrum analyzer is set to 14 ms of sweep time and 30 kHz of resolution bandwidth.

Gyrotron radiation has a finite linewidth which can be attributed to both intrinsic noise sources, such as shot noise, flicker noise, velocity noise, thermal agitation noise, as well as extrinsic technical noise sources such as fluctuations in the operating parameters [6, 16]. Linewidth and lineshape measurements of the gyrotron are very important in the applications utilizing the frequency shift by scattering, for example, radar [17] and collective Thomson scattering for plasma diagnostics [18]. DNP/NMR also requires narrow linewidth to hit exact frequency in the EPR spectrum for maximum signal enhancement in NMR. In order to measure the linewidth of the gyrotron over a large dynamic range (60 dBc and 75 dBc), we used a low noise harmonic mixer. The input power to the mixer was increased within 10% of the LO power. In that configuration, we could lower the amplification of the noise floor down to -80 dBm from -70 dBm in the spectrum analyzer.

Three separate frequency sweeps lasting 0.1 s each have been overlapped in Fig. 6 to estimate the linewidth and lineshape. The power level of the center frequency (299.87854 MHz) in the IF domain is about -3.5 dBm. Therefore, the 60 dBc linewidth at -63.5 dBm is measured to be around 1.0 MHz, and the 75 dBc linewidth at -78.5 dBm is estimated to be 4.3 MHz, close to the detection limit set by the phase noise of the local oscillator circuit (about 0.3 MHz at 60 dBc). After converting the power scale into a linear ratio relative to the peak power, the full-width at half-maximum (FWHM) linewidth is calculated to be 72 kHz. The true FWHM linewidth of the gyrotron could be narrower because the linewidth in the heterodyne measurement is the convolution of the LO with the gyrotron. These result shows that gyrotrons have good spectral characteristics and are suitable for many practical applications.
Figure 6 Spectral linewidth of the 140 GHz gyrotron in the IF domain, in which the LO frequency was locked at 19.99400 GHz and its 7th harmonic was mixed with the gyrotron frequency. For this measurement, the spectrum analyzer was set to 10 kHz of resolution bandwidth and 100 ms of sweep time. Sweeping was delayed by 10 s to wait until the frequency was stabilized after the onset of radiation.

VI. DISCUSSION

In this article, we reviewed the present status of compact CW gyrotrons being developed at MIT for use in DNP/NMR spectrometers to obtain significant enhancements in spin spectra from biological samples. To date, 140 GHz and 250 GHz gyrotrons have been employed in DNP experiments at 200 MHz and 380 MHz spectrometers at MIT. A 460 GHz gyrotron operating at the second electron cyclotron harmonic, to be employed with an NMR spectrometer (700 MHz), has been demonstrated in stable operation for 24 hours. These gyrotrons are capable of operating at a relatively low beam voltage of 12 kV and producing CW power of a few watts at very high frequency with good power stability. With good spectral and spatial resolution, these tubes should have important applications in research in physics, chemistry, biology, materials science and medicine. For example, high output power in this frequency range enables the construction of an imaging system, in conjunction with appropriate beam splitter and detector arrays that avoids a time consuming scanning process. With a flexible quasi-optical dielectric guiding system [15] to direct these strong submillimeter-wave beams to the portion containing cancer cells in a human body, effective therapy could be established [19, 20]. Other promising applications utilizing these powerful CW submillimeter-wave sources are open for synergetic collaborations.

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REFERENCE


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