# High-Order Multipole Excitation of a Bound Electron

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The nonlinear resonant response of a bound electron to a time-varying spatially inhomogeneous electric field was studied experimentally. By use of the artificial atom "geonium" (an electron bound in a Penning trap), we observed up to ninth-order multipole (pentacosiododecapole) coherent excitation of the electron's magnetron motion, and up to third-order (octupole) excitation of the cyclotron motion. Also, by applying two fields simultaneously, we have observed coherent stimulated Raman excitation of the electron's motion.

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The interaction of an electromagnetic field with a medium is, in general, nonlinear. There are three common origins of the nonlinearity [1–3]: (1) the interaction of the charges with the spatial inhomogeneity of the excitation fields, (2) the anharmonic binding of the medium's charges, and (3) the interaction of the medium with the magnetic component of the field. A number of resonant nonlinear effects have been studied, for example, harmonic generation, multiwave mixing, and multiphoton absorption. These nonlinear effects have almost exclusively relied on electric dipole transitions (with notable exceptions [4]), and the origin of the nonlinearities falls primarily into the second category. The first nonlinear mechanism, which corresponds to excitation of higher-order multipole transitions, is less common. Although multipole excitation is observed in some atomic and nuclear physics experiments, the excitation and detection are usually incoherent. We have studied this nonlinear mechanism by using an artificial atom [5]: an electron bound in a Penning trap [6–8]. This has provided for the first time a means to excite and detect, coherently, high-order multipole transitions, and observe high-order subharmonic generation. We have also observed several degenerate and nondegenerate nonlinear parametric processes which have been discussed by Kaplan and Ding [2,3]. Our observations complement the body of work on coherent harmonic excitation and multiwave mixing in atoms.

An electron bound in a Penning trap can be thought of as the artificial atom "geonium" [6–8]. The "nucleus" of geonium (the trap) is created with the electrodes (and fields) shown schematically in Fig. 1. A static voltage $V_0$ applied between the end caps and ring forms a potential (in spherical coordinates)

$$\Phi(r,\theta,\phi)=V_0\sum_{n=0}^{\infty}C_{2n}(r/d)^{2n}P_{2n}(\cos\theta)\,, \tag{1}$$

where the $P_{2n}$ are the Legendre polynomials and the $C_{2n}$ are constants. Here $d=r_0/\sqrt{2}=3.54$ mm is a characteristic trap size, with $r_0$ the ring internal radius. The term labeled by $C_2$ forms a harmonic well along the $\hat{z}$ axis. Our trap is designed to have $C_2=0.477$, and $C_4 \simeq C_6 \simeq 0$ [9]. Applying a voltage $V_g$ to two additional guard electrodes [10] allows $C_4$ to be varied; in practice, $|C_4|<5\times10^{-5}$. Although $|C_4|$ was small, it remains the dominant electrostatic perturbation to our harmonic potential term. The leading perturbation to an applied homogeneous magnetic field $B_0\hat{z}$ is a magnetic quadrupole bottle [6–8] of order $B_2/B_0=7\times10^{-5}$ cm$^{-2}$ due to magnet imperfections and the copper electrode diamagnetism. $B_0$ was measured to have a minimum long term drift rate of $|(dB_0/dt)/B_0|=7\times10^{-7}$ h$^{-1}$, and for times on the order of 1 min, field fluctuations of $\Delta B_0/B_0 \simeq 3\times10^{-7}$ for $B_0 \simeq 0.1$ T.

The motion of a nonrelativistic electron bound in an ideal Penning trap ($C_n=0$ for $n>2$, $B_0$ homogeneous) is composed of three independent modes [6–8]. The axial ($z$) motion is harmonic with amplitude $r_z$ and frequency $\omega_z=(2C_2qV_0/md^2)^{1/2}$ where $q/m$ is the electron's charge-to-mass ratio. In the radial ($x$-$y$) plane the motion is two superimposed circular motions: the magnetron motion of radius $r_m$ and frequency $\omega_m$ corresponding to the $\mathbf{E}\times\mathbf{B}$ drift of the electron, and the cyclotron motion of radius $r_c$ and frequency $\omega_c'=\omega_c-\omega_m$ where $\omega_c=qB_0/mc$. For $V_0=-10.45$ V and $B_0=0.1$ T we obtain $\omega_z/2\pi=61.5$ MHz, $\omega_m/2\pi=615$ kHz, $\omega_c'/2\pi=3.08$ GHz, and in general we maintain $\omega_c' \gg \omega_z \gg \omega_m$. The axial energy is damped [$\Gamma_z$(measured)$\simeq 20$ s$^{-1}$] by connecting a resonant tuned circuit to one of the end cap electrodes (analogous to a two-level atom in a resonant optical cavity) [6–8,11]. The magnetron motion is meta-

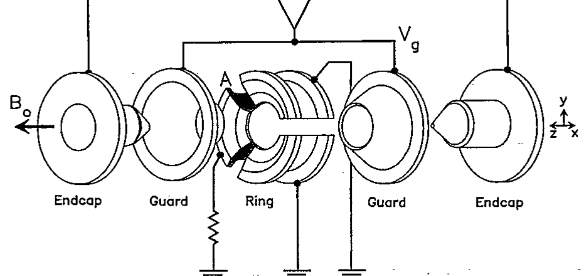


FIG. 1. A schematic diagram of the Penning trap used in the experiment. The electrodes have been separated along the $z$ axis to show details.

 

stable. However, its amplitude can be damped at a variable rate of $0 \le \Gamma_m \le \frac{1}{2}\Gamma_z$ to a theoretical limit of $r_m = 2[(\omega_m/\omega_z)\langle r_z^2\rangle]^{1/2}$ [where $\langle r_z^2\rangle^{1/2}$ is the thermalized ($T=4$ K) axial amplitude] by the use of sideband cooling [6-8]. The cyclotron energy is radiatively damped at the free space rate modified by the presence of the trap and secondary electrodes [12] [$\Gamma_c$(measured)$\simeq 0.1$ s$^{-1}$ (1 s$^{-1}$) for $B_0 \simeq 0.1$ T (1.4 T)]. Three main factors lead to deviations from the ideal motion: (1) anharmonic terms in the electrostatic potential ($C_n \ne 0, n>2$), (2) relativistic effects, and (3) inhomogeneity of the magnetic field [6-8,13]. These cause the electron's modes of motion to be slightly anharmonic and weakly coupled together.

A single electron is loaded into the trap using established techniques [6-8,11]. The frequencies $\omega_z, \omega_m, \omega_c'$ are measured by applying oscillating drive potentials to different electrodes and observing a resonant response of the electron's motion. For the axial motion, the electron's response is observed by measuring the image currents induced in one end cap using a phase-sensitive heterodyne detector (analogous to phase sensitive detection of atomic resonance fluorescence) [6-8,11]. We chose to detect the magnetron or cyclotron excitations via their coupling to the axial motion [6-8]. Specifically, the observed axial frequency depended upon the magnetron amplitude due to the electrostatic $C_4$ term, and it depended on the cyclotron amplitude due to the slight magnetic field inhomogeneity $B_2$. For example, for a magnetron excitation ($r_m = 0 \to r_{m0}$), $\Delta\omega_z/\omega_z \simeq -3C_4(r_{m0})^2/2C_2d^2$. The shift $\Delta\omega_z$ is effectively observed by monitoring the correction voltage of an electronic servomechanism that locks the axial frequency to an external reference [6-8]. If, in addition to the driven oscillation (at frequency $\omega_d$) there is a free (or thermally excited) oscillation at $\omega_m$, then an interference beat note at $\omega_d - \omega_m$ will be observable in the correction voltage [7,14]. If a second coherent drive field is applied with frequency $\omega_0$, then three beat notes can be observed with frequencies $\omega_d - \omega_m$, $\omega_0 - \omega_m$, and $\omega_d - \omega_0$. The magnetron and cyclotron motions are excited by applying a drive voltage to sector $A$ of the ring electrode (Fig. 1). To estimate the ($x$-$y$) drive field the electrode geometry is approximated by a box with sides of length $(l_x, l_y, l_z) = 2d(\sqrt{2}, \sqrt{2}, 1)$. One side of the box perpendicular to $\hat{x}$ (representing sector $A$) is assumed to be at a potential $V_d$ with respect to the other sides. This model gives the resulting potential (in the $x$-$y$ plane)

$$\Phi_d(x,y,0) = V_d \sum_m \sum_n \alpha_{m,n} (x/d)^m (y/d)^n , \qquad (2)$$

with $\alpha_{m,n}$ constants [15].

A quantum description of the nonlinear excitation involves calculating amplitudes for processes like those in the insets in Figs. 2 and 3. However, in our experiment the mean occupation numbers of the electron's quantized motion were much larger than 1, so a classical model suffices [2,3]. When excited by a homogeneous field, the magnetron and cyclotron motions can be treated as one-

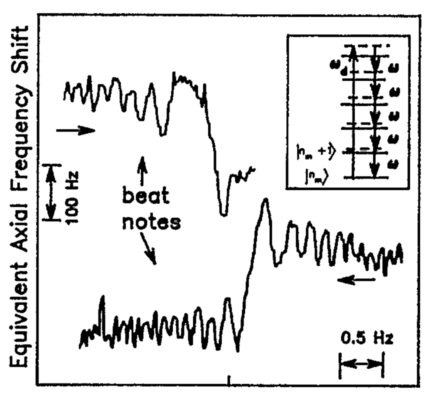


FIG. 2. The subharmonic response of order $\omega_d/\omega_m \simeq 5$ of the center-of-mass magnetron motion of a cloud of approximately seventy electrons. Baselines for the two traces have been offset for clarity. The arrows show the direction the drive frequency was swept (at a rate of 0.1 Hz/s). The axial frequency shifts due to the magnetron excitation (the vertical steps) and the beat note between the free oscillation at $\omega_m$ and the subharmonically excited oscillation at $\omega_d/5$ are observed. For this figure the sideband cooling was off and the magnetron amplitude had been initialized in a large orbit. The inset shows a representation of this coherent process for particular quantum levels.

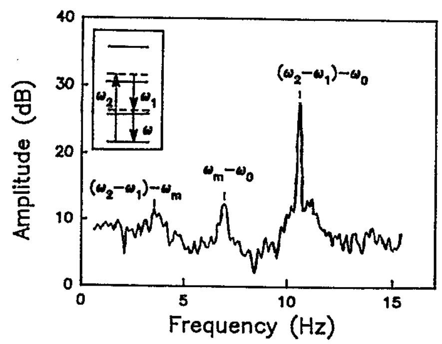


FIG. 3. The Fourier transform of the response of the magnetron motion of a single electron to drive fields at frequencies $\omega_0$, $\omega_1$, and $\omega_2$. The magnetron free oscillation is at frequency $\omega_m$, and the driven oscillations are at $\omega_0 = \omega_m - 2\pi(7$ Hz) and $\omega_2 - \omega_1 = \omega_m + 2\pi(4$ Hz), with $\omega_1 = \omega_m + 2\pi(97$ Hz). Three interference beat notes are observed. For short averaging times, $\tau < 10$ s, the amplitudes and widths of the three beat notes were comparable. For longer averaging times ($\tau = 100$ s is shown here) the beat note responses between the driven and free oscillations (left two peaks) broaden and decay because of the magnetic field noise affecting the free oscillation. The beat between two driven oscillations (right peak) is unaffected (other than an amplitude modulation noise pedestal) and its width remains limited by the resolution of the dynamic signal analyzer (FFT). When measured with a phase detector, this peak showed that the nonlinear response was phase coherent with the drives. The inset shows a representation of this stimulated Raman response at $\omega_2 - \omega_1$.

dimensional harmonic oscillators. For an inhomogeneous drive field this is no longer true, but the main effect of the field inhomogeneity is to modify somewhat the strength and phase of the force. We therefore assume that the $x$ components of the magnetron and cyclotron motions obey the approximate equations of motion (ignoring mode coupling)

$$\ddot{x}+\Gamma_j\dot{x}+\omega_j^2x[1+\xi(t)+\beta_2(x/d)^2+\beta_4(x/d)^4+\cdots] \\ =a_jF_d[\alpha_0+\alpha_1(x/d)+\alpha_2(x/d)^2+\cdots]\cos(\omega_dt)\,, \tag{3}$$

where $F_d=qV_d/md$, $\alpha_0=0.19$, $\alpha_1=0.35$, $\alpha_2=0.29$, and $\alpha_3=0.13$ [from Eq. (2)], and $j$ refers to either the cyclotron or magnetron motion. This equation neglects the $y$ and $z$ dependence of the force. However, since the $\alpha_i$ are only approximate, this further simplification should not significantly affect the results. For the magnetron motion $\omega_j=\omega_m$, $a_j=\omega_m/(\omega_m-\omega_c')\simeq-\omega_m/\omega_c'$, and $\beta_i\propto C_{i+2}$. For the cyclotron motion $\omega_j=\omega_c'$, $a_j\simeq\omega_c'/(\omega_c'-\omega_m)\simeq1$, and the $\beta_i$ arise from the relativistic mass shift [13]. The term $\xi(t)$ is a multiplicative noise term [16] which, in our experiment, is caused primarily by the magnetic field fluctuations. The magnetic component of the drive fields [category (3) of the introduction] can be shown to be negligible here. Equation (3) holds also for the center-of-mass motion of small clouds of electrons (cloud size $\ll d$); experimentally, both small clouds and single electrons were used.

Equation (3) is solved by assuming solutions of the form $x(t)=A(t)\cos[\omega t+\phi(t)]$ where $A(t)$ and $\phi(t)$ are slowly varying. Nearly resonant excitation ($\omega\simeq\omega_j$) of the motion can occur when $\omega_d\simeq n\omega_j$, $n$ an integer, in which case $\omega=(1/n)\omega_d$. This corresponds to subharmonic excitation (i.e., frequency division). It can be caused by (1) the electron's motion in the inhomogeneous drive field [$\alpha_i$ in Eq. (3)], and/or (2) the anharmonic motion of the electron [$\beta_i$ in Eq. (3)], as noted in the introduction. The first is due to electric multipole transitions of frequency $n\omega_j$. The second is due to dipole transitions of frequency $n\omega_j$ which arise from mixing of the quantum oscillator levels due to the anharmonicity. Since $\beta_{n-1}\ll n^2\alpha_{n-1}/\alpha_0$ for our experiment, Eq. (3) can be used to show that the first mechanism dominates. Parametric excitation ($n=2$) has been observed previously in traps for the axial motion of a single electron [11] and clouds of electrons [17–19].

If we neglect $\beta_i$ with $i>2$, Eq. (3) reduces to Duffing's equation [3] driven by a nonlinear force. Subharmonic response of order $n$ exhibits both hysteresis and $n$ stable phases of response relative to the phase of the drive [2,3]. In addition, from Eq. (3), a steady state response requires [for $\xi(t)=0$] [3,20]

$$\left|\frac{a_j\alpha_{n-1}F_d}{2^{n-1}\omega_jd}(A/d)^{n-2}\right|\geq\Gamma_j\,. \tag{4}$$

Therefore $F_d$ and $A$ must exceed certain threshold values $F_{dt}$ and $A_t$. The origin of these thresholds is that the energy supplied by the drive at $\omega_d$ to the oscillation at $\omega_d/n$ is dependent upon the amplitude $A$ of that oscillation and must overcome the damping. No such threshold exists for harmonic excitation (frequency multiplication), $\omega_d\simeq\omega_j/m$. The initial amplitude $A_t$ necessary for subharmonic response can be generated by a resonant homogeneous drive ($\omega_d\simeq\omega_j$), a thermal drive, or a free oscillation. When frequency noise is included [$\xi(t)\neq0$], $\Gamma_j$ in Eq. (4) is replaced by an effective damping rate which depends upon the form of the noise [16], and the frequency sweep rate of the drive. However, the ratios of Eq. (4) for different orders of $n$ should be insensitive to $\Gamma_j$.

Subharmonic excitation of the magnetron motion was observed for $n=1$–$9$ on samples of up to about seventy electrons when the magnetron amplitude was initialized in a large orbit. Both the frequency shift and modulation of the axial frequency (by the beat notes) were observed as the magnetron amplitude resonantly increased. This is seen in Fig. 2 for $n=5$, where the beat note is due to the interference between the magnetron free oscillation and the subharmonically excited driven oscillation.

With a single electron the threshold values for subharmonic response for $n=2$, 3, and 4 were measured with the initial magnetron amplitude cooled to its minimum value (set by the sideband cooling). Independently, we determined the magnetron amplitude at its sideband cooling limit by varying $C_4$ and measuring the resulting change in $\omega_m$ [8,14]. This gave $r_m=220\pm140$ $\mu$m, or $r_m=(55\pm35)r_{\rm theo}$, where $r_{\rm theo}$ is the theoretical cooling limit. This disagreement with theory is consistent with measurements made by other groups, and its origin is not known [7,8,14]. We measured the threshold values $V_d(n=2)=6$ $\mu$V, $V_d(n=3)=300$ $\mu$V, and $V_d(n=4)=6$ mV, for $\omega_m/2\pi=615.0$ kHz ($B_0=0.1$ T, sideband cooling off, sweep rate 0.1 Hz/s). From these measurements and Eq. (4), we calculated $\alpha_1/\alpha_2=1.6\pm1.0$ and $\alpha_1/\alpha_3=1.0^{+1.7}_{-0.8}$. These agree with the values given by the electrostatic model [Eq. (2)] within the measurement uncertainty. Similar results were found for $\omega_m/2\pi=47.34$ kHz ($B_0\simeq1.4$ T).

The origin of the subharmonic response at $n=3$ was experimentally investigated. This response could arise from the excitation of the octupole moment of the magnetron orbit, or from a weakly allowed dipole transition due to the $C_4$ anharmonic term in the trap electrostatic potential. Changing the guard voltage allowed $C_4$ to be varied in small steps from $C_4<5\times10^{-5}$ to $>4\times10^{-4}$. No change in the required threshold drive strength was observed. If the subharmonic response had been due to the anharmonic component of the magnetron motion, this threshold drive strength would be proportional to $|C_4^{-1}|$.

The more general resonance condition [21] $\omega_d=(n/m)\omega_m$, $m,n\neq1$ was also investigated. We observed resonant excitation when $\omega_d-\frac{2}{3}\omega_m$ (for $V_d=3$ mV, $\omega_m/2\pi=615.0$ kHz, and the same conditions as above), which corresponds to three-photon excitation of the quadrupole resonance.

For two drive fields we observed stimulated Raman excitation of the magnetron motion when $\omega_2 = q\omega_1 = r\omega_m$ with ($q=1$, $r=1,2,3$) and ($q=1,2,3$, $r=1$). These resonances could be observed either with the sideband cooling (damping) on or off, and typical detunings $\Delta = \omega_1 - \omega_m = 2\pi(1 \text{ to } 150 \text{ Hz})$. Resonant enhancement of the response was observed as $\Delta$ was decreased. The beating between these excitations and the free magnetron oscillation was observed as well as the anharmonic pulling of the magnetron frequency. When another driven oscillation near $\omega_0 \simeq \omega_m$ was generated, three beat notes were observed, two between the driven and free oscillations, and one between the two driven oscillations near $\omega_m$ (see Fig. 3). The observation of these beat notes allows the magnetron responses (linear or nonlinear) to be measured in a continuous phase sensitive manner [20].

We observed subharmonic excitation of order $n=2$ and 3 for a single electron's cyclotron motion ($\omega_c'/2\pi \simeq 3$ GHz). Since $\omega_c' \gg \omega_m$, magnetic field drift and noise caused significantly more dephasing of the cyclotron motion than that of the magnetron motion; this resulted in poor quantitative agreement between the cyclotron results and the simple theory. When two drive fields were applied with frequencies $\omega_2$ and $\omega_1$, stimulated Raman excitation of the cyclotron motion of a single electron was observed for $\omega_2 - \omega_1 \simeq \omega_c'$ with $\omega_1 = \omega_c' + \Delta$. Resonant enhancement of the response was observed as the detuning $\Delta$ was decreased; typically $\Delta/2\pi = 100$ to 600 kHz. Magnetic field noise prevented us from observing beat notes between the different cyclotron excitations.

In summary, we have observed, coherently, high-order nonlinear resonant excitation of a single bound electron's motion as well as that of the center-of-mass motion of a cloud of electrons. These effects were observed in the limit where the nonlinearity was dominated by the multipole interaction between the electron and the drive field. Initial comparisons with a simple one-dimensional theory show quantitative agreement for excitation of the magnetron motion. Detailed quantitative comparisons between experiment and theory requires better magnetic field stability, better estimates for the shape of the drive fields (the $\alpha_i$) and orbit amplitudes, as well as the extension of Eq. (3) to three dimensions. In addition, the effects of mode coupling and noise-induced fluctuations of the mode frequencies must be included in the nonlinear analysis, especially because they can inhibit very high-order excitation. Experimentally, it should be possible to greatly reduce the magnetic field noise, which in turn should facilitate high-order subharmonic excitation of the cyclotron motion. If very high-order subharmonic response can be observed $\omega = \omega_d/n$ with $n \simeq 10^4$, the system could have applications in frequency metrology [3,22]. In general, this system might be useful in studies of the effect of noise on nonequilibrium phase transitions [16], as well as chaotic behavior in the transition from classical to quantum dynamics.

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