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"BOLOMETRIC" TECHNIQUE FOR THE rf SPECTROSCOPY OF STORED IONS*

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A simple technique is proposed in which rf transitions between suitable energy levels of an atomic or molecular ion, a member of a stored, radiatively thermalized ion gas, may be detected by monitoring the translational temperature of the ion gas. An experimental study of the cyclotron resonance of an electron gas confined in a Penning trap demonstrates its usefulness, and possible applicability to spin resonance.

The rf spectroscopy of stored ions in the past has relied on collision schemes in which polarized beams of alkali atoms, photons, etc., are used to polarize the ions and also monitor the ion polarization thus realized by means of spin-dependent simultaneous ion-loss processes.[1] Such schemes are of necessity rather complex. It therefore appears useful to propose here a relatively simple "bolometric" scheme, in which the translational temperature of a stored ion-gas cloud is monitored while the atomic or molecular ions composing it are undergoing rf transitions between suitable, e.g. hfs, rotational, electronic, etc., energy levels. The scheme presupposes, of course, the existence of sufficiently strong relaxation processes connecting the levels in question with the ion motion. Experimentally observed ion lifetimes, days for electrons[1] and hours for protons,[2] and the high degree of thermal isolation of the ion gas realizable indicate that quite weak relaxation processes may be sufficient. Specifically, one may think of electric or magnetic interactions between the ions and of the effects of motion through inhomogeneous magnetic and electric fields. The latter effect (Majorana flops) will, of course, be most pronounced if the (internal) transition frequency ω_i to be detected coincides with a characteristic frequency of the translational periodic motion in the trap ω_t. For $\omega_i \neq \omega_t$ it is still possible to "assist" the relaxation process by modulating the field gradient at $|\omega_i - \omega_t|$, whereby the atom oscillating through the pulsating gradient is made to see a sideband of the oscillatory translational motion falling on ω_i. Other relaxation interactions are undoubtedly conceivable. It is further assumed that collisions between the stored ions establish very quickly a thermal distribution of ion energies. It is then possible to cool the ions by coupling their disordered axial motion to a suitably tuned LC circuit which also acts as the thermometer.[2,3] The scheme may be considered to be the simplest embodiment of the ion storage-collision technique in rf spectroscopy on which the earlier experiments mentioned above are also based. Rather then going into a discussion of conceivable hypothetical embodiments of the proposed scheme, we decided on an experimental study of a particularly suitable system. To this end the detection of the cyclotron resonance in an electron gas at approximately room temperature stored in a trap based on the principle of the Penning gas discharge was chosen.[1,3]

A block diagram of the apparatus used in these experiments is shown in Fig. 1. Electrons from

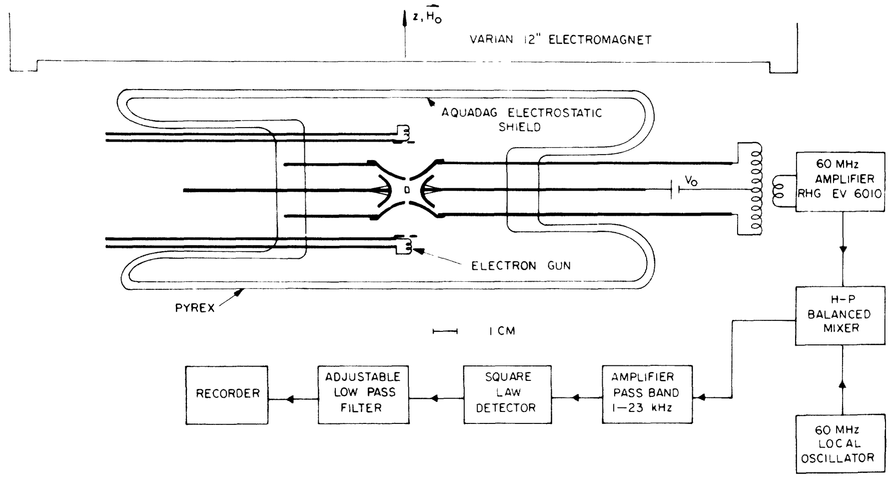

FIG. 1. A cross-sectional view of the electron trap and a block diagram of the electronics used in the bolometric detection of the electron cyclotron resonance. The small rectangular slot in the center "ring" electrode is a microwave window. The vacuum manifold with its 15-liter/sec Vacion pump and GE 22GT210 triggered discharge gauge is connected to the front section of the glass envelope which was cut away to expose this view.

a hot tungsten filament are injected through a hole in one end cap of a quadrupole ion trap, presumably knocking off slow electrons from the residual gas inside it. Alternatively, the electron beam is reflected upon itself. The resulting e-e collisions transfer the parallel energy of some electrons into transverse motion, temporarily trapping them, the coupling to the tank circuit finally providing stable containment. The gold electrodes are carefully machined hyperbolas of revolution with $2r_0 = 1.25$ cm and $2z_0 = 0.76$ cm. When the center ring electrode is held at a positive dc potential V_0 with respect to the two end electrodes or caps, they provide a harmonic well of depth $\sim\frac{1}{2}V_0$ for electrons along the z axis. Confinement in the perpendicular x-y plane is achieved by applying a static magnetic field H_0 along the z axis of symmetry. The motion of the electrons is a synthesis of three separate motions. First, there is the harmonic oscillation parallel to the z axis with frequency ω_z which is unaffected by the magnetic field. Second, there is the cyclotron motion with frequency $\omega_c = eH_0/mc$. This motion is slightly perturbed to $\omega_c - \omega_m$ by the radial electric fields also giving rise to the third, the magnetron motion, which is a slow drift of the center of the cyclotron orbits about the z axis. The frequency ω_m of this motion is related to the two other frequencies by the relation $2\omega_m^2 - 2\omega_m\omega_c + \omega_z^2 = 0$. The stored electrons are thermalized by radiatively coupling them to the room temperature thermal reservoir. The following simple model exhibits many of the important features of this process: Consider a harmonically bound ion with charge q and mass m oscillating with frequency ω_z along the z axis, which is perpendicular to the faces of an infinite parallel-plane capacitor in which it is confined. The capacitor is assumed to have negligible capacitance; its plates, separated by a distance d, are connected together by a large resistor R. The oscillating ion induces a current $i = qv_z/d$ to flow through the resistor, where v_z is the instantaneous z component of velocity of the ion. The Joule heating of the resistor exponentially damps the ion's motion with a time constant $\tau_{z0} = md^2/q^2R$. The energy of n harmonically bound ions moving with random phases is damped at the same rate. The effect of replacing the infinite-plane capacitor by a quadrupole ion trap, $d = 2z_0$, is to increase τ_{z0} by approximately 30%. The condition of negligible capacitance is satisfied when the z-oscillation frequency of the ions equals the resonant frequency of the tank circuit. For the purpose of obtaining a measure of the ion temperature the noise voltage across the tank circuit is amplified, square-law detected, and filtered yielding a dc voltage proportional to the noise temperature T_t of the tank circuit. When the ions are externally heated or cooled, the tank-circuit temperature and hence the dc level are changed accordingly.

While in the general case there may be many translational and internal degrees of freedom associated with the ion gas under study, in order to bring out the most essential characteristics of our technique, it is sufficient to restrict ourselves here to a system exhibiting only two, namely, the heated and the monitored, degrees of freedom. In our specific experimental example they are realized by the "perpendicular" cyclotron motion in the x-y plane and the longitudinal oscillatory "z" motion. The sensitivity of this technique for detecting temperature changes in the perpendicular motion depends on the number of ions and the coupling times τ_{t0}, τ_{zt}, and τ_{pz}, the latter referring to transfer between perpendicular and longitudinal motion due to e-e collisions. It may be assumed $\tau_{zt} = \tau_{z0}$, where τ_{zt} refers to transfer from the z motion to the tuned circuit rather than the reservoir (R). To find the temperature increase of the tank circuit due to perpendicular-motion heating, one has to analyze the system sketched in Fig. 2. The characteristic times listed above refer to the initial rates with which the respective process occurs following an instantaneous perturbation of the corresponding component of this coupled system, say, the p motion. Neglecting residual heating due to nonspecific causes, one has to solve the following set of equations for energy transfer between the various energy reservoirs:

$$c_p dT_p/dt = -c_p(T_p - T_z)/\tau_{pz} + \text{heating},$$

$$c_z dT_z/dt = +c_p(T_p - T_z)/\tau_{pz} - c_z(T_z - T_t)/\tau_{zt},$$

$$k dT_t/dt = +nc_z(T_z - T_t)/\tau_{zt} - k(T_t - T_0)/\tau_{t0}.$$

Here c_p is the single-ion heat capacity in the perpendicular motion, c_z is that in the z motion, k is Boltzmann's constant, and n is the number of ions. T_p, T_z, T_t, and T_0 are the corresponding temperatures of the heated level, of the z motion, the tank circuit, and the infinite heat reservoir. For the simple case where one sweeps through resonance slowly compared with τ_{zt}, the system is essentially at equilibrium and one can set all of the time derivatives equal to zero. Solving the equations for the temperature of the tank circuit in terms of T_p, one finds for the "sensitivity"

$$\eta \equiv (T_t - T_0)/(T_p - T_0) = (n/n_c)(1 + n/n_c)^{-1}.$$

Here the critical ion number n_c and the dimensionless quantities α and β are defined by

$$n_c \equiv (1+\alpha)/(\alpha\beta), \quad \alpha \equiv c_p\tau_{zt}/c_z\tau_{pz},$$

$$\beta \equiv c_z\tau_{t0}/k\tau_{zt}.$$

For $n \gg n_c$, the sensitivity η becomes independent of n, approaching unity, and the noise temperature of the tuned circuit is that of the ions. For $n \simeq n_c$, T_t will be intermediate between T_p and T_0 and $\eta < 1$. On the basis of the foregoing it is possible to measure the sensitivity η experimentally and also to determine the number of ions in the trap by the following method: The tank circuit is heated with white noise to a known temperature T_h for a time long compared with $\tau_{zt} \gg \tau_{pz}$. This insures that the ions and the tank circuit are at the same temperature. When the heating is suddenly terminated, the temperature of the tank-circuit noise thermometer will rapidly decrease with an approximate time constant of τ_{t0} from T_h to T_t while the ion temperature practically remains T_h. The relation for η may now be applied:

$$(T_t - T_0)/(T_h - T_0) = \eta \equiv (n/n_c)(1 + n/n_c)^{-1}.$$

The subsequent slow decay of T_t with a time constant of $\sim 2\tau_{zt} \gg \tau_{t0}$ need not be considered. The temperature scale of the noise thermometer was shot noise calibrated by running the emission current from the filament operating in a temperature-limited mode through one-half of the tank circuit. The base-line temperature is assumed to be equal to 300°K since no appreciable extraneous heating of the electron gas has been observed. The above methods are quite satisfactory for $T_z \gtrsim 300$°K. However, when the equilibrium temperature of the ions is to be kept small compared with the equivalent input noise temperature of the amplifier, small changes in ion temperature are difficult to observe. In such instances it is advantageous to use parametric postmultiplication of the ion temperature. This is done by modulating the dc well depth at $2\omega_z$. By using rf pulses of constant amplitude and du-

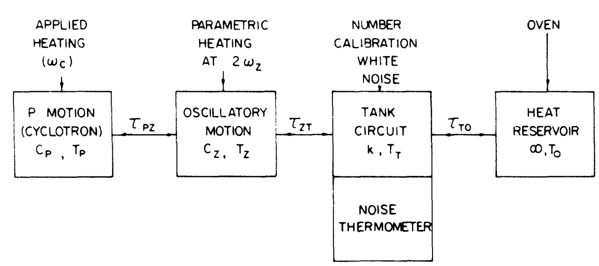

FIG. 2. Diagram of the coupled system of heated and monitored motions (degrees of freedom) underlying the "bolometric" technique for the rf spectroscopy of stored ions. The heat reservoir is physically represented by the resistance R shunting the (lossless) tank circuit.

ration $\ll\tau_{pz}$, one can repeatedly multiply the temperature by a constant factor and hence obtain a noiseless gain. In detection schemes relying on particle counting, it may be possible to translate ion temperature doubling by the rf resonance into particle ejection from the trap even at $T_z \approx 4°\mathrm{K}$.

We have observed absorption of energy by electrons at frequencies ω_m, ω_z, $2\omega_z$, and ω_c. Figure 3(a) shows some typical data taken near the cyclotron resonance. The data were taken at $V_0 \approx 12$ V and a magnetic field of 7.7 kG, with $\omega_z = 60$ MHz, $\tau_{t0} = 0.3\times10^{-6}$ sec, and $n = \frac{1}{3}n_c = 1.0\times10^5$ electrons. The magnetic field was held constant and the frequency of the microwave field swept through the resonance. The temperature scale is based on noise-thermometer readings. The sidebands are due to the ω_m motion of the electrons through the inhomogeneous microwave field. Analogous motion-induced sidebands occur in the "assisted" relaxation mechanism mentioned earlier. A rf power-dependent displacement of all the lines towards lower frequencies is observed. It is presumed due to the second-order Doppler shift, which is proportional to the temperature of the electrons. From the Doppler shift of the ω_c peak, which amounts to 6 ± 1 parts in 10^8, the temperature increase of the electrons at resonance is estimated to be $230\pm40°\mathrm{K}$, which is in agreement with the noise-thermometer data. The uncertainty in the base-line temperature is due to an uncertainty in how much excess noise from the input circuit of the first stage of the detection amplifier appears across the tank circuit. The characteristic times for momentum transfer from p to z motion, $\tau_{pz} < 5$ msec, and for radiation damping, $\tau_{zt} = 0.1$ sec, were deduced from transients of the electron temperature induced by pulse excitation of cyclotron and z motion. A measure of the thermal isolation of the electron gas was obtained by increasing the frequency of the tuned circuit by a factor of 5 in order to minimize the thermal contact of the electrons with it. This increases the observed τ_{z0} (from all causes) to ~2 min. No increase is observed when the temperature of the electron gas, thus isolated when initially at $\simeq T_0$, is measured minutes later. Although the proposed parametric amplification scheme is more novel than useful for detecting temperature changes of ions at room temperature, its great potential for low-temperature applications makes its verification important. Figure 3(b) shows the cyclotron peak with and without parametric amplification for $T_0 \simeq 300°\mathrm{K}$. Data were taken by replacing the recorder with a voltage-to-frequency converter and a multichannel analyzer.

The decay of the electron number is not exponential, making it impossible to quote a simple lifetime. For a typical run and an initial number n_0 of $6n_c$ there remains approximately $1n_c$ after $\frac{1}{2}$ h, $0.2n_c$ after 10 h, and $0.05n_c$ after 100 h. The pressure was measured using a General Electric 22GT210 triggered discharge gauge. The vacuum system is approximately half stainless steel and half Corning 7740 Pyrex in terms of surface area. From the results of Davis[4] on residual gas in stainless vacuum systems with Vacion pumps, the predominant constituents of the residual gas were assumed to be CO, H_2, N_2, and He in equal amounts. The sensitivity of the gauge is then approximately 0.5 A/Torr.[5] The gauge current was normally about 1×10^{-11} A corresponding to a pressure of 2×10^{-11} Torr.

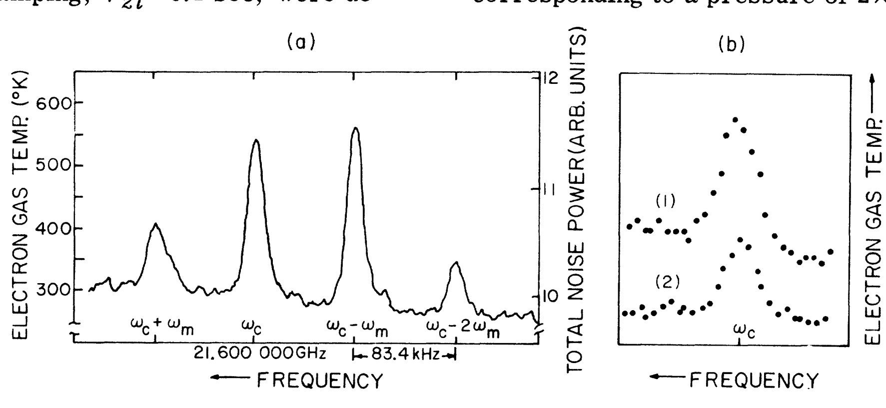

FIG. 3. (a) Temperature increase of the electron gas as the exciting frequency is swept through the cyclotron resonance. The temperature scale is correct to approximately 50°K (see text). The slight slope of the base line is due to losses from the initially injected electron sample during the duration of the sweep which was approximately 4 min. (b) The cyclotron line with (1) and without (2) parametric postmultiplication of the electron-gas temperature. Off-resonance points of (1) and (2) refer to room temperature.

In conclusion, our results as a whole indicate also that the rf-saturated spin resonance of the electrons in the cloud may be detectable by means of "assisted" Majorana flops connecting it with the cyclotron motion, the latter being strongly connected to the temperature-monitored z motion. Experiments towards this goal are in progress in our laboratory.

We wish to acknowledge the collaboration of Dr. T. S. Stein in the last phases of the work. Mr. Jake Jonson made major contributions in the construction of the Penning-trap tube.

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[1]H. G. Dehmelt, Advances in Atomic and Molecular Physics (Academic Press, Inc., New York, 1967), Vol. 3, p. 53.

[2]D. A. Church and H. G. Dehmelt, to be published.

[3]H. G. Dehmelt, Bull. Am. Phys. Soc. 7, 470 (1962), and 8, 23 (1963).

[4]W. D. Davis, Trans. Nat. Vacuum Symp. 9, 363 (1962).

[5]W. J. Lang, J. H. Singleton, and D. P. Eriksen, J. Vac. Sci. Technol. 3, 338 (1966).

EMISSION OF PULSE TRAINS BY Q-SWITCHED LASERS*

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Maxwell's equations and the two-level density-matrix equations are integrated numerically under conditions which simulate the operation of a Q-spoiled laser. The final output intensity pattern takes the form of a train of sharp pulses separated by the round-trip time.

The phenomenon of laser mode locking which results in periodic short-pulse emission has been demonstrated in a variety of experiments.[1] Particularly dramatic results have been obtained with lasers which are Q switched by means of bleachable dyes.[2] Recently, it has been demonstrated by the use of two-photon fluorescence techniques that short-pulse emission can result when Q switching is accomplished by other methods such as a rotating prism.[3] The possibility that such short pulses could arise as fluctuations of a Gaussian random process has been examined.[4] There is, on the other hand, considerable laboratory experience to indicate that pulsed lasers or lasers Q switched by electro-optic methods can spontaneously emit narrow pulses at round-trip time intervals.[5] In this communication, we report that the emission of such periodic pulse trains from lasers employing Q switches other than saturable absorbers can be predicted theoretically.

We take as the equations which describe the amplification of the laser field in a homogeneously broadened medium[6] the following, which are based on Maxwell's equations and the conventional two-level density-matrix equations:

$$E(z,t)=E^+(z,t)e^{i\omega(t-\eta z/c)}+E^-(z,t)e^{i\omega(t+\eta z/c)}+\text{c.c.},\tag{1a}$$

$$\rho(z,t)=\rho^+(z,t)e^{i\omega(t-\eta z/c)}+\rho^-(z,t)e^{i\omega(t+\eta z/c)},\tag{1b}$$

$$\frac{\eta}{c}\frac{\partial E^\pm}{\partial t}\pm\frac{\partial E^\pm}{\partial z}=-\tfrac{1}{2}\sigma\rho^\pm,\tag{1c}$$

$$\partial\rho^\pm/\partial t+T_2^{-1}\rho^\pm=-T_2^{-1}nE^\pm,\tag{1d}$$

$$\partial n/\partial t+T_1^{-1}(n-n_0)=2\sigma(\rho^+E^{+*}+\rho^-E^{-*}+\text{c.c.}).\tag{1e}$$

In Eq. (1a) the laser field $E(z,t)$ is expressed as the sum of two running waves moving in opposite directions inside the laser cavity. Both E^+ and E^- are complex. The normalization of E is such that $2|E|^2$ is a measure of photon intensity. The carrier frequency ω is taken for convenience as the laser transition frequency. The variables ρ^+ and ρ^- are proportional to the polarizations induced by the respective E waves, and n is the population difference. Also, η is the index of refraction, σ is the absorption cross section at the line center, T_1 and T_2 are the longitudinal and transverse relaxation times, and n_0 is a constant associated with the pumping rate. In writing Eq. (1e), we have neglected terms of the form $\rho^+E^{-*}\times e^{-2ikz}$ and $\rho^-E^{+*}e^{2ikz}$. In order to treat such terms, it is necessary to expand ρ^+, ρ^-, and n in power series in e^{2ikz}. This procedure, involving