The effect of the laser mode structure on the sideband spectrum of stored ions

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The optical absorption spectrum of ions oscillating in a three-dimensional harmonic trap is calculated for laser excitation. The effect of the mode structure of the laser beam employed, in particular the case of the TEM_{00} mode used in most experiments is considered. The spectrum contains many sidebands, besides the central transition frequency, generated by *all* combinations of the ion oscillation frequencies in the three orthogonal directions of motion, even when the laser is directed along the trap axis or is incident in the radial plane, and even when perfect symmetry of the trapping fields is assumed. It is shown that this coupling between the axial and radial frequencies is due to the Gaussian mode structure of the laser beam. In laser spectroscopy of ions trapped in a real radio frequency (rf) trap, energy transfer between the ion motional degrees of freedom is therefore caused by the mode structure of the laser beam and, as previously assumed, by asymmetries of the pseudopotential well and for more than one ion in the trap by ion space charge fields. The ion motion coupling due to the laser mode structure is of particular significance during the initial phases of laser cooling experiments when comparatively hot ions sample a distance comparable to the waist of the collimated laser beams and when this distance is of the order of the optical wavelength.

I. INTRODUCTION

The need for the reduction of the first and second order Doppler shifts, which limit ultrahigh resolution spectroscopic measurements and their application to the metrology of time, has been ever present in the history of spectroscopy. However presently substantial progress is being made to nearly eliminate all Doppler effects by radiation pressure cooling of atoms[1] and sideband cooling of ions trapped in electromagnetic traps.[2] These techniques are well established for atoms[3] and ions.[4] The optical excitation experiments involving laser cooling of trapped ions[5] have reached the highest quality factors $Q=\nu/\Delta\nu$, where ν is the transition frequency and $\Delta\nu$ is the observed linewidth of the trapped ions; however microwave experiments on trapped ions even without cooling have also reported similar Q values earlier.[6] In the laser experiments involving sideband cooling of trapped ions one takes advantage of the first order Doppler effect which manifests itself in an interesting way. When the ion motion is restricted to excursions of the order of λ and when the motion of the ions is harmonic, the absorption or emission spectrum of the ions displays not only the spectral line at the transition (central) frequency but also at a series of frequencies on both the lower and upper sides of the transition frequency. These additional features are known as sidebands. Such a sideband spectrum was first observed and studied[7] in the microwave region on the spectrum of helium ions stored in a rf quadrupole trap[8,9] and later on by others.[10] More recently sidebands have been observed[11,12] and theoretically investigated[13] in the optical region.

In the work reported here the influence of the mode structure of the laser beam on the sideband spectrum was studied. The mode structure is also of interest for ion cooling when, during the initial phases of cooling a small cloud of trapped ions sample the waist of the narrowly collimated laser beam. The calculation is made using the formalism of the correlation functions[14–16] since the stored ion cloud samples different laser fields at different times and the perturbation is spread out in frequency. In this calculation the Gaussian nature of the laser beam, used for the excitation of the optical spectrum of the ions stored in a rf quadrupole trap, is taken into account and the important case of the TEM_{00} mode is considered in detail.

II. CALCULATION

The optical spectrum of the oscillating ions (in a rf quadrupole trap) can be calculated using the correlation function formalism. The power spectrum $P(\omega, \omega_0)$ (equivalently the transition rate) associated with the frequency $\omega_0=(E_m-E_k)/\hbar$ is proportional to the Fourier transform of the correlation function

$$P(\omega,\omega_0) \propto \int_{-\infty}^{\infty} G_{mk}(\tau) e^{-i\omega_0\tau} d\tau, \tag{1}$$

where E_m and E_k are the energies of the levels participating in the transition and $G_{mk}(\tau)$ is the correlation function defined as the ensemble average of the matrix elements of the perturbation Hamiltonian in the stationary perturbation approximation. $G_{mk}(\tau)$ is formally written as

$$G_{mk}(\tau) = \langle H_{mk}(t') H_{km}(t) \rangle, \tag{2}$$

where $\tau=(t-t')$ and the matrix elements of the perturbation Hamiltonian H involve the electric field of the radiation inducing the electric dipole transitions. Since different

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ions are at different locations, the electric field seen by them at a given time will be different, which implies that the electric field is implicitly dependent on the motion of the ions.

The motion of the ions in a trap is determined by the operating parameters of the trap and consists of the large amplitude macromotion at frequencies ω_x, ω_y, and ω_z with the small amplitude micromotion at the driving frequency Ω superimposed on the macromotion. In the following, only the dominant mode of the oscillation at the macromotion is considered. This assumption is justified since the micromotion has a much smaller amplitude than the macromotion. The micromotion has a negligible amplitude around the bottom of the pseudopotential well where the cool ions reside. The macromotion of the ions along the three symmetry axes of the trap is given by

$$\overline{X}=x\sin(\omega_x t+\phi_x),$$
$$\overline{Y}=y\sin(\omega_y t+\phi_y) \quad (3)$$
$$\overline{Z}=z\sin(\omega_z t+\phi_z).$$

The amplitudes x, y, and z together with the phase angles ϕ_x, ϕ_y, and ϕ_z determine the exact trajectory assumed by a particular ion in the trap. Because of the rotational symmetry of the trapping fields in a rf trap, the oscillation frequencies in the radial plane are equal and $\omega_x=\omega_y$ holds.

Next it is assumed that the electromagnetic radiation, inducing the transitions among the energy levels of the oscillating ions, is in the TEM_{00} mode and that the laser beam is propagating along the z axis. In this case the electric field of the radiation is given by

$$E=E_0 e^{-(\overline{X}^2+\overline{Y}^2)/\Delta^2}\cos(k\overline{Z}-\omega t), \quad (4)$$

where $k=2\pi/\lambda$ is the wave number, ω is the angular frequency of the radiation, and Δ is the spot size (or beam radius) of the laser beam. Generally Δ varies along z, the coordinate along the propagation direction. The minimum spot size is the waist. In the following we assume Δ to be constant in the trap which is equivalent to collimating the laser beam throughout the trapping volume. In terms of the electric field of the laser radiation the correlation function can be written as

$$G_{mk}(\tau)=A_{mk}\int_0^{2\pi}\int_0^{2\pi}\int_0^{2\pi}\int_{-x_m}^{x_m}\int_{-y_m}^{y_m}\int_{-z_m}^{z_m} \times d\phi_x d\phi_y d\phi_z dx\,dy\,dz f(x,y,z)E(t)E(t'), \quad (5)$$

where $f(x,y,z)$ is the distribution function characterizing the ion oscillation amplitudes in the trap, x_m, y_m, z_m are the maximum ion amplitudes (for hot ions these are the trap dimensions in the orthogonal directions), and A_{mk} is a constant determined by the transition matrix elements. Using Eqs. (3) and (4) in Eq. (5) for the correlation function and integrating over the phase angles one obtains

$$G_{mk}(\tau)=CA_{mk}\int_{-x_m}^{x_m}\int_{-y_m}^{y_m}\int_{-z_m}^{z_m}\Bigg[dx\,dy\,dz\,f(x,y,z) \times\cos\omega t\sum_{l,m,n}^{\infty} I_l^2\left(\frac{x^2}{2\Delta^2}\right)I_m^2\left(\frac{y^2}{2\Delta^2}\right)J_n^2(kz) \times e^{-x^2/\Delta^2}e^{-y^2/\Delta^2}\cos(2l\omega_x\tau)\cos(2m\omega_y\tau) \times\cos(n\omega_z\tau)\Bigg], \quad (6)$$

where C is a constant of integration that also includes all other constants. I_l and I_m are modified Bessel functions (of the first kind) of order l and m, J_n is the Bessel function (of the first kind) of order n, and l, m, and n are integers.

The absorption spectrum is obtained by inserting Eq. (6) into Eq. (1). The result is

$$P(\omega,\omega_0)=P_0(\omega_0)\Bigg(\int_{-x_m}^{x_m}\int_{-y_m}^{y_m}\int_{-z_m}^{z_m}dx\,dy\,dz\,f(x,y,z) \times\sum_{l,m,n} I_l^2\left(\frac{x^2}{2\Delta^2}\right)I_m^2\left(\frac{y^2}{2\Delta^2}\right)J_n^2(kz)e^{-x^2/\Delta^2} \times e^{-y^2/\Delta^2}\delta[\omega-(\omega_0\pm 2l\omega_x\pm 2m\omega_y\pm n\omega_z)]\Bigg), \quad (7)$$

where $P_0(\omega_0)$ includes all constants.

III. DISCUSSION

The power spectrum given by Eq. (7) clearly contains many spectral lines, besides the central line at the transition frequency ω_0, which are generated by the different combinations of ω_x, ω_y, and ω_z. This power spectrum for the Gaussian beam in the TEM_{00} mode has more spectral lines than the spectrum produced when a uniform plane wave is absorbed. A plane wave does not couple the ion motion frequencies when incident along the z axis and yields only z sidebands. Consider for a plane wave a laser beam whose waist in the trap is very large compared to the ion oscillation amplitudes. In this case $(x^2,y^2/2\Delta^2)\ll 1$, and all modified Bessel functions are zero except the zeroth order ones. Thus only $l=0$ and $m=0$ terms survive in the spectrum and the resulting spectrum, which is also the spectrum of a harmonic oscillator in one dimension, is

$$P(\omega,\omega_0)=P_0(\omega_0)\Bigg(\int_{-x_m}^{x_m}\int_{-y_m}^{y_m}\int_{-z_m}^{z_m}dx\,dy\,dz\,f(x,y,z) \times\sum_n J_n^2(kz)\delta[\omega-(\omega_0\pm n\omega_z)]\Bigg), \quad (8)$$

where $P_0(\omega_0)$ is a constant.

The intensities $I_{l,m,n}$ of different spectral lines, in the power spectrum given by Eq. (7) for the Gaussian TEM_{00} mode of radiation, are obtained according to

$$I_{l,m,n}=\int_{-x_m}^{x_m}\int_{-y_m}^{y_m}\int_{-z_m}^{z_m} dx\,dy\,dz\,f(x,y,z)$$
$$\times[e^{-x^2/2\Delta^2}I_l(x^2/2\Delta^2)]^2[e^{-y^2/2\Delta^2}I_m(y^2/2\Delta^2)]^2$$
$$\times[J_n(kz)]^2, \tag{9}$$

where the integrals are evaluated numerically. With an arbitrary ion amplitude distribution function, the evaluation of Eq. (9) is difficult. In laser cooling the distribution function changes continuously with temperature. However, for special cases, for instance if the ion motion amplitude distribution is uniform, meaning that all amplitudes are equally probable up to a maximum value, the integrals for the three variables uncouple and a numerical integration can be readily performed. A uniform distribution is present after ion production, when the effect of the laser mode structure is large. The same uniform amplitude distribution was also assumed for the calculation of the plane wave absorption.

Figure 1(a) depicts the sideband spectrum in the case that a laser beam in the TEM_{00} mode and at a wavelength of $\lambda=268$ nm is employed. The transition chosen as an example is the intercombination line ($^1S_0\rightarrow{}^3P_1$) of the boron ion. The lifetime of the upper $^3P_1$ state is about 100 ms and hence long enough to display a natural linewidth that is orders of magnitude smaller than the calculated smallest sideband spacing. The operating parameters of the trap are compiled in Table I. The x, y integrals are evaluated with $x_m=y_m=\Delta$, which implies that the laser beam is collimated to overlap with the ion motion maximum amplitudes. Only those lines which have an intensity greater than 1% of the intensity of the central line are shown, and only the sidebands that occur within a range of 3 MHz of the central line are depicted. There are many more sidebands when a larger bandwidth is considered. Also only the high frequency side of the spectrum is shown since the complete spectrum is symmetrical about the central frequency. Figure 1(b) depicts, for comparison, the sideband spectrum due to a uniform wave propagating in the z direction. Comparing the two figures, it is evident that the effect of the mode structure is to generate additional sidebands also involving the ion oscillation frequencies in directions orthogonal to the direction of the incident radiation. Some of the sidebands occur at the same frequencies as the sidebands due to the uniform plane wave absorption. This is due to the integer combinations of the macromotion frequencies involved and is part of the degeneracy of the spectrum. It is evident from the figures that the intensities of the sidebands generated by the ion oscillation frequencies perpendicular to the z axis are significantly smaller than the intensities of the sidebands generated by the motion along the z axis. These sidebands are actually amplitude modulated sidebands, since they are caused by the exponential decrease of the electric field of the laser radiation with distance from the z axis, which is characteristic of Gaussian beams. Nonetheless, the presence of these additional sidebands implies a coupling between the motion of the ions in the z direction and along the two other orthogonal directions.

TABLE I. Parameters used to calculate the sideband spectrum for TEM_{00} mode and plane wave absorption.

Axial dimension (z_0)	0.07 cm
Radial dimension (r_0)	0.10 cm
rf trapping frequency ($\Omega/2\pi$)	15 MHz
Axial oscillation frequency ($\omega_z/2\pi$)	1.0 MHz
Radial oscillation frequency ($\omega_{x,y}/2\pi$)	0.7 MHz

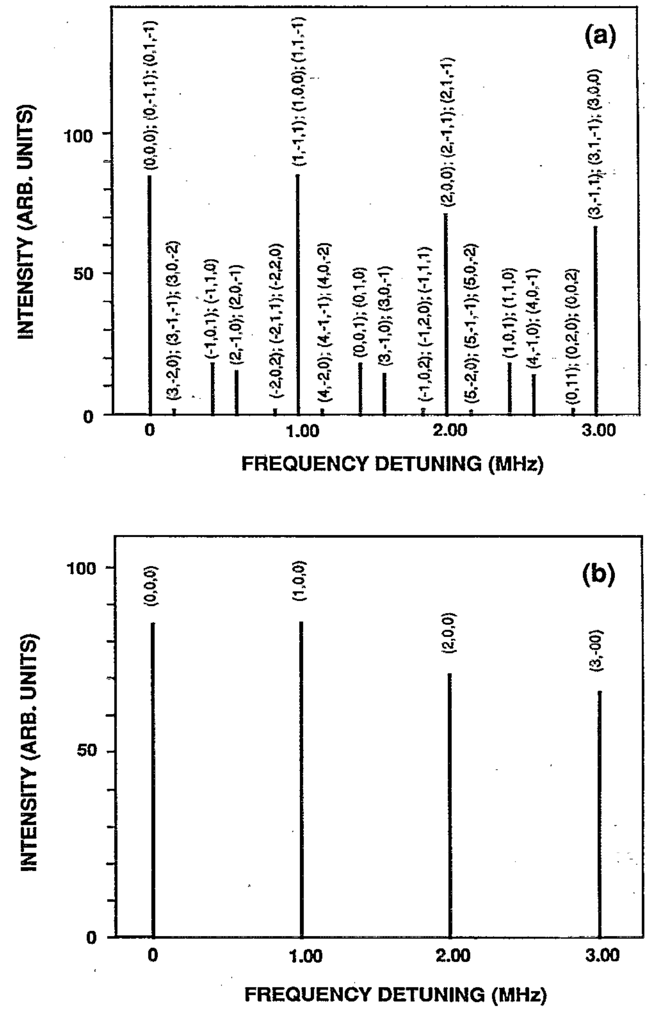

FIG. 1. (a) Calculated sideband spectrum due to TEM_{00} mode laser beam absorption along the z axis of the trap. (b) Calculated sideband spectrum due to uniform plane wave absorption along the z axis of the trap. In both plots relative intensities are displayed versus the frequency detuning from the line center. The numbers in the parentheses above the lines indicate the order of the sideband corresponding to the integers n, l, and m. A uniform amplitude distribution of the ion motion was assumed.

IV. CONCLUSIONS

It was shown that the laser mode structure couples the axial and radial ion oscillation frequencies even in the case when the laser is collimated along one of the symmetry axes of the trap. This coupling is also present when ideally the ions execute independent harmonic motions in the axial and x, y directions, and manifests itself in a sideband spectrum which contains *all* harmonics of the z and x, y macromotions. If one uses the relative intensities of the sideband spectrum to determine the temperature of stored ions, this mode structure effect can be significant, particularly when relatively hot ions are investigated with a tightly collimated laser beam.

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[1] T. W. Hänsch and A. L. Schawlow, Opt. Commun. **13**, 68 (1975).
[2] D. J. Wineland and H. G. Dehmelt, Bull. Am. Phys. Soc. **20**, 637 (1975).
[3] See, for example, in the special issue "Laser Cooling and Trapping," edited by S. Chu and C. Wieman, J. Opt. Soc. Am. B **6**, 2023 (1989).
[4] J. C. Bergquist, R. G. Hulet, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **57**, 1699 (1986).
[5] J. J. Bollinger, D. J. Heinzen, W. M. Itano, S. L. Gilbert, and D. J. Wineland, Phys. Rev. Lett. **63**, 1031 (1989).
[6] R. Blatt, H. Schnatz, and G. Werth, Phys. Rev. Lett. **48**, 1601 (1982).
[7] H. A. Schuessler, Appl. Phys. Lett. **18**, 117 (1971).
[8] H. S. Lakkaraju and H. A. Schuessler, J. Appl. Phys. **53**, 3975 (1982).
[9] H. A. Schuessler, E. N. Fortson, and H. G. Dehmelt, Phys. Rev. **187**, 5 (1969).
[10] M. Jardino, F. Plumelle, M. Desaintfuscien, and J. L. Duchene, *Proceedings of the 38th Annual Frequency Control Symposium* (IEEE, New York, 1986), 84CH2062-8, p. 431; L. S. Cutler, C. A. Flory, R. P. Giffard, and M. D. McGuire, Appl. Phys. B **39**, 251 (1986).
[11] J. C. Bergquist, W. M. Itano, and D. J. Wineland, Phys. Rev. A **36**, 428 (1987).
[12] F. Dietrich, J. C. Bergquist, W. M. Itano, and D. J. Wineland, Phys. Rev. Lett. **62**, 403 (1989).
[13] D. J. Wineland, W. M. Itano, J. C. Bergquist, and R. G. Hulet, Phys. Rev. A **36**, 2020 (1987).
[14] C. P. Slichter, *Principles of Magnetic Resonance* (Harper & Row, New York, 1963).
[15] F. G. Major and J. L. Duchene, J. Phys. (Paris) **36**, 953 (1975).
[16] J. H. Van Vleck and P. L. Huber, Rev. Mod. Phys. **49**, 939 (1977).