Physica Scripta. Vol. T70, 53–56, 1997

Four-Quantum RF-Resonance in the Ground State of an Alkaline Atom

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Received November 16, 1995; accepted March 25, 1996

Abstract

The multiple-quantum resonances in Zeeman structure of the ground state of Potassium atoms can be observed in Earth magnetic field under intense RF excitation. They complicate the magnetic resonance spectrum and can lead to errors in the magnetic field measurement by Potassium magnetometer. On the other hand, the highest-order 4-quantum resonance $F = 2$, $m_F = 2 \Leftrightarrow m_{F'} = -2$ shows up special features which make it attractive for application in Earth field magnetometry. Unlike any other single- or multiple-quantum resonances, the frequency of this one is strictly linear with the magnetic field strength and practically independent of the driving RF-field amplitude. Besides, this resonance has the greatest resolving power and can be very easy selected on the back-ground of the other broadened resonances. The problem of the magnetic resonance in 8-level system has been solved numerically without any restriction on the RF-field intensity. The experimentally recorded spectra match the theory.

1. Introduction

The theme of the paper goes back to the times of golden age of Optical Pumping in 1950–1960. In those days a lot of refined studies of radio-spectroscopy of atoms was undertaken. In particular, multiple-quantum transitions were observed and interpreted by A. Kastler team when studying RF-spectra of optically pumped sodium atoms [1] and by P. Kusch using atomic-beam technique applied to potassium atoms [2]. These and many other investigations were summarized in the substantial paper [3] of J. Winter (see also the review of A. Bonch-Bruevich and V. Khodovoi [4]). Later on, the interest in multiple-quantum processes shifted from RF-domain to optical range where double-quantum transitions in counter-propagating light beams became very popular because of their ability to suppress Doppler broadening of spectral lines [5]. As to RF-domain, multiple-quantum resonances did not find any further application in research or metrology in spite of their obvious advantage: an n-quantum transition is n times narrower than the single-quantum one (in the limit of low driving field power). But this advantage is heavily depreciated by a rather strong dependence of the resonance frequency on the driving field power. Indeed, the use of the multiple-quantum processes from the most general point of view seems to be inexpedient because of necessity to perturb the system by much stronger driving field.

Nonetheless, it looks like we have found a particular case of RF-induced multiple-quantum transition in the ground state of alkaline atom which could be of interest for the low magnetic field metrology. We mean four-quantum resonance $m_F = 2 \Leftrightarrow m_F = -2$ of the atom with nuclear moment $I = 3/2$ (see Fig. 1). Two features of this transition attract attention: (i) the energy of the unperturbed transition is strictly linear *vs.* the magnetic field strength H unlike any other transition and (ii) the frequency of this transition seems to be almost unaffected by the driving field H_1 again unlike all other multiple-quantum transitions. The latter feature follows from estimates based on a perturbation approach [4]: under the influence of the field H_1 both states $m_F = \pm 2$ are expected to shift almost equally. This prediction needs to be confirmed by accurate calculation which is the main aim of the following consideration.

In the past multiple-quantum processes were theoretically analyzed mainly in the framework of the perturbation approach – as higher terms of power expansion of the transition probability. Apart from the well known Bloch's exact solution of the resonance problem in two-level system only three-level system interacting with single- or double-frequencies field has been thoroughly analyzed in many papers (see for instance [6–9] and references therein). In principle, using the approximation of rotating fields the exact analytical stationary solution can be obtained for any

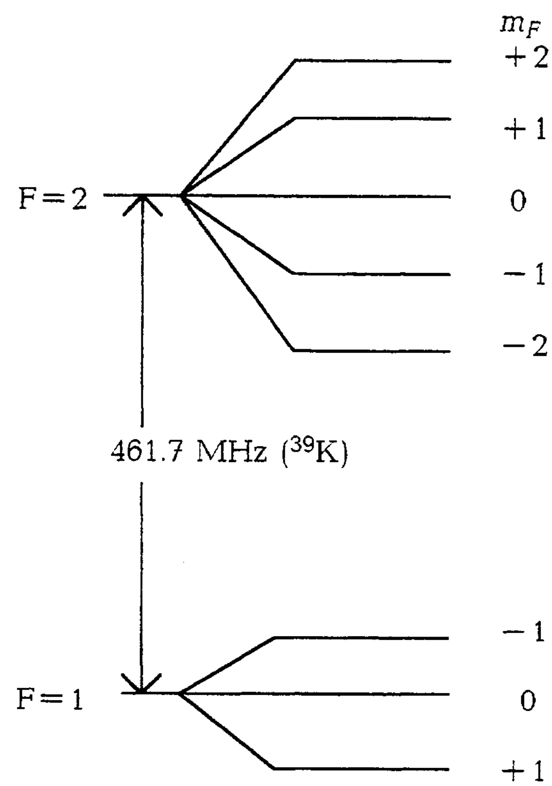

Fig. 1. Scheme of the ground state magnetic splitting of the alkaline atom ^{39}K with nuclear moment I = 3/2.

k-level system. It is noteworthy, however, that for $k \geqslant 4$ it is highly troublesome job with extremely cumbersome and practically uninterpretable results. But nowadays there exist powerful computers and advanced software which make it possible to get easily numerical or even analytical solution of many-level problem valid for any power of the driving field. Below we present the result of computer simulation of 4-quantum RF resonance in optically pumped alkaline atoms.

Formulation of the problem

The frequencies $f_{m,\,m-1}$ of the RF-transitions between adjacent magnetic sublevels m and $m-1$ of the ground $^2S_{1/2}$ state obey the well-known Breit–Rabi formula. They are presented below as the power expansion over the magnetic field strength H:

$$f_{2,\,1} = a_+ H - 3bH^2 + cH^3 - \cdots$$

$$f_{1,\,0} = a_+ H - bH^2 - cH^3 + \cdots$$

$$f_{0,\,-1} = a_+ H + bH^2 - cH^3 - \cdots$$

$$f_{-1,\,-2} = a_+ H + 3bH^2 + cH^3 + \cdots$$

There is a set of four lines within the state $F = 2$, and of two lines for $F = 1$:

$$f_{1,\,0} = a_- H + bH^2 - cH^3 - \cdots$$

$$f_{0,\,-1} = a_- H - bH^2 + cH^3 + \cdots$$

The coefficients $a_\pm$ are very near to $\pm 7\,\mathrm{GHz/T}$:

$$a_\pm = f_i \pm f_j \cong \pm f_j$$

where $f_i = g_i \mu_B/h$, $f_j = (g_j - g_i)\mu_B/(4h)$, g_i and g_j are nuclear and electron g-factors, μ_B and h are the Bohr magneton and the Planck constant.

$$b = f_j^2/\Delta\nu_{hfs}$$

$$c = 6bf_j/\Delta\nu_{hfs},$$

$\Delta\nu_{hfs}$ being the hyperfine splitting of the ground state.

Spectrum of the RF-induced transition, at sufficiently low power of the driving field H_1 when the RF-field induced width $f_j H_1$ of the transitions remains much smaller than the frequency difference between the adjacent lines, should comprise the six above lines. The same is valid with respect to intrinsic width Γ_{ik} of the line. Under such conditions the RF-field interacts with the multiple-level system as with an ensemble of independent two-level systems. But as the field is increased, the lines become broader, their resonance positions shift, and additional two-photons peaks appear with the resonance frequencies $\omega_j^{(2)} \approx (\omega_{j+1} - \omega_{j-1})/2$, where ω_k is the frequency (energy) of the level k. With further increase of H_1 similar triple- and four-quantum transitions appear within sublevels of $F = 2$ state. It should be mentioned that the four-quantum transition has never been experimentally observed so far, probably, because it could not be distinguished on the background of the overlapping broadened neighbouring lines. To make it clear it is necessary to provide great sharpness of the spectrum, i.e., a larger ratio of the lines splitting to the line width. In the following calculation we will assume intrinsic line width to be extremely small – 1 Hz – consistent with our recent experimental results [10].

Let us consider the optically pumped atomic vapour in so called M_z-configuration. It means that the vapour is being pumped by circular polarized resonant light beam along the magnetic field H and magnetic resonance is being observed as a change of pumping light absorption κ:

$$\kappa = \approx \sum p_i w_i \tag{1}$$

where p_i and w_i are the population and the absorption probability of sublevels number i. So, the problem is to calculate the population distributing under the action of optical pumping and a driving RF-field.

We will seek a steady-state solution of the equation for the density matrix ρ_{jk} completed with phenomenological terms to describe the optical pumping and relaxation processes. The equation for off-diagonal terms:

$$i\hbar\, \partial\rho_{jk}/\partial t = [H_0 + V, \rho]_{jk} - i\hbar(\Gamma\rho)_{jk} \tag{2}$$

where Hamiltonian H_0 (with diagonal matrix) describes atoms in a constant magnetic field, the operator V takes into account effect of the RF-field H_1 and the matrix $\Gamma\rho$ describes the relaxation of the coherency ρ_{jk} due to thermal processes and optical excitation. We assume, for simplicity, that in the absence of light excitation all elements ρ_{jk} relax with the same rate Γ_0. Pumping light shortens the life time of atoms and thus additionally broadens (by Γ_k) the level k. The coherency ρ_{jk} relax with the rate $\Gamma_{jk} = \Gamma_0 + (\Gamma_j + \Gamma_k)/2$.

The eq. (2) neglects the coherency transfer in the course of optical pumping which corresponds to the buffer gas optical pumping.

Dealing with populations $p_j \equiv \rho_{jj}$ it is necessary to add to the eq. (2) terms describing the optical pumping. In the absence of pumping all population relax to the same value 1/8. Under optical pumping the atom at the level j is excited to the state μ and after spontaneous decay goes to the sublevel k of the ground state with the probability $B_{kj} = b_{kj} I_h$ proportional to the pumping intensity I_p. Finally, the equations for the populations ρ_{jj} become:

$$\begin{aligned}\partial\rho_{jj}/\partial t = (i\hbar)^{-1}[H_0 + V, \rho]_{jj} - \rho_{jj}(\Gamma_0 + \Gamma_j)\\ + \Gamma_0/8 + \sum \rho_{kk} B_{jk}\end{aligned} \tag{3}$$

The probabilities b_{kj} and w_i are listed in the paper of Franzen and Emslie [11].

In rotating field approximation $V = V \exp(i\omega t)$, where ω is driving field frequency. Seeking for the steady-state solution, we will find coherencies $\rho_{jk}(t)$ in the form $\rho_{jk}(t) = \rho_{jk} \exp[i\omega(j-k)t]$. We will also keep in mind that an RF-field does not produce microwave coherencies between sublevels belonging to the different hyperfine states. It has also nonzero matrix elements V_{jk} for $j - k = \pm 1$ only. Representing ρ_{jk} as $\rho_{jk} = x_{jk} + iy_{jk}$, we can reduce the eqs (2) and (3) to the set of 34 linear algebraic equations for 8 populations (steady-state conditions imply $\partial\rho_{jj}/\partial t = 0$) and 26 values x_{jk} and y_{jk}.

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* $a_+/a_- = (7.004\,666/-7.008\,639)\,\mathrm{GHz/T}$; $b = 106.327\,\mathrm{GHz/T^2}$; $c = 9681\,\mathrm{GHz/T^3}$.

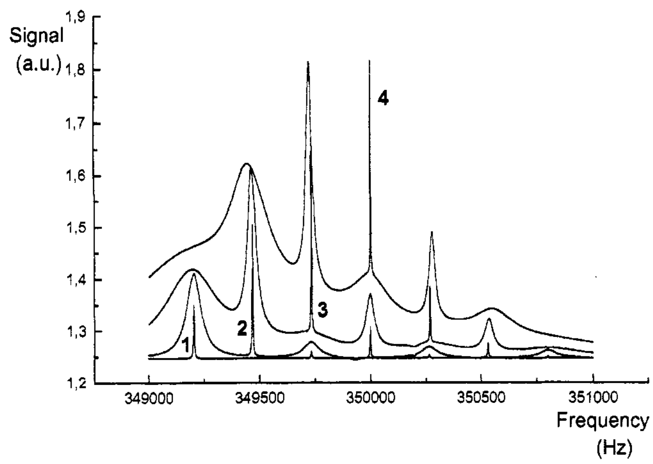

Fig. 2. RF-spectra of the ground state double-resonance signal of ^{39}K at four different values of the driving field H_1 strength (in units of $g = f_j H_1$): (1) $g = 1$; (2) $g = 20$; (3) $g = 80$; (4) $g = 190$.

Results

A realistic situation of $^{39}K^*$ in the average earth field of about $50\,\mu T$ has been simulated. The pumping light intensity was chosen so that the main (strongest) single-photon resonance $f_{2,1}$ became 1.5 times broader as compared with its "dark" width Γ_0. It corresponds (more or less) to the condition of getting an maximum resolution for the single-photon RF-spectrum [12].

The set of 34 equations has been solved numerically and a number of graphs $\kappa(\omega)$ has been plotted for different values of H_1. Figure 2 shows a family of graphs $\kappa(\omega)$ for $f_j H_1$ from 1–190 in units of Γ_0 which was assumed to be 1 Hz. The H_1 strength in each case was selected so that to maximize sharpness of each n-quantum resonance in succession from $n = 1$ to $n = 4$. Only very weak traces of transitions within sublevels of the state $F = 1$ can be found on two of four spectra. Figure 3 presents in more details the RF-spectrum in the vicinity of four-photon resonance. The rather flat back-ground of the resonance is related to the deeply saturated two-photon resonance $m_F = 1 \Leftrightarrow m_F = -1$ at almost the same frequency. Figure 4 displays the maximal steepness $S_m(f_j H_1) = max[\partial S/\partial\omega]$, where S is signal strength. The signal steepness characterizes the accuracy of the resonance peak location, playing the leading role in evaluation of the resonance line for application. Figure 4 compares the steepness of the main ordinary resonance with that of four-quantum one. One can see that the steepness of the last one is about 7 times greater. Finally, Fig. 5 shows the resonance shift induced by driving field for the single- and four-quantum lines. For each resonance the field H_1 was chosen in the vicinity of its optimal value. One can see that field induced shifts are of the same order being in fact negligible.

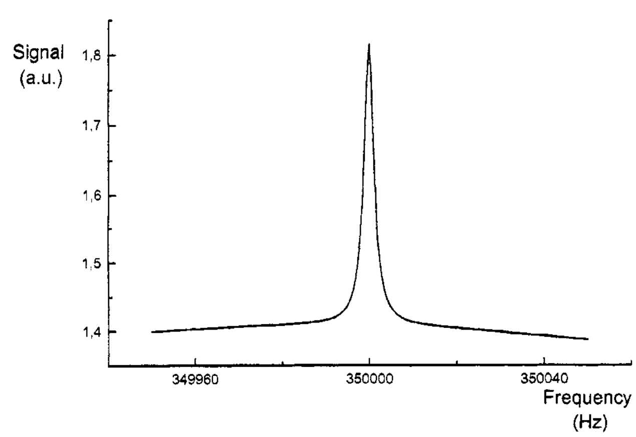

Fig. 3. The four-quantum resonance contour at $g = 190$.

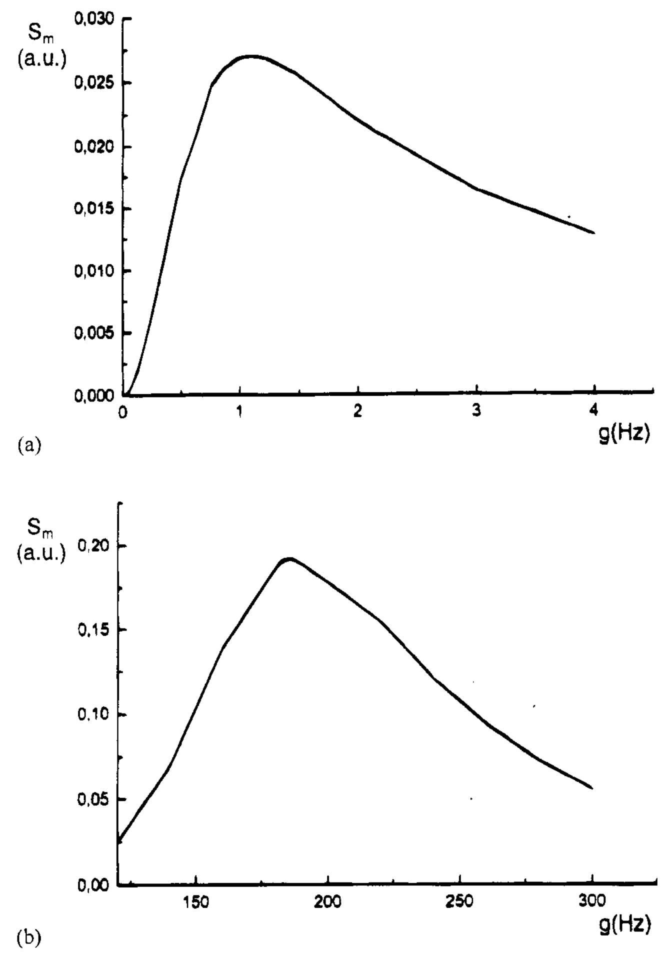

Fig. 4. The maximal steepness of the single-quantum (a) and four-quantum (b) resonances *vs.* driving field strength (arbitrary units).

Conclusion

The results of detailed computer simulation support our initial supposition about attractive features of the four-quantum resonance: its quality (frequency discriminating ability) surpasses that of the ordinary resonance and its position is not affected by fairly strong driving field. So, it seems that four-quantum resonance can compete with the single-photon one in magnetometric application. One should remember the point about strictly linear dependence of this resonance frequency on magnetic field strength. And the last point is that under optimal driving field this resonance can be easy selected from adjacent broadened resonances.

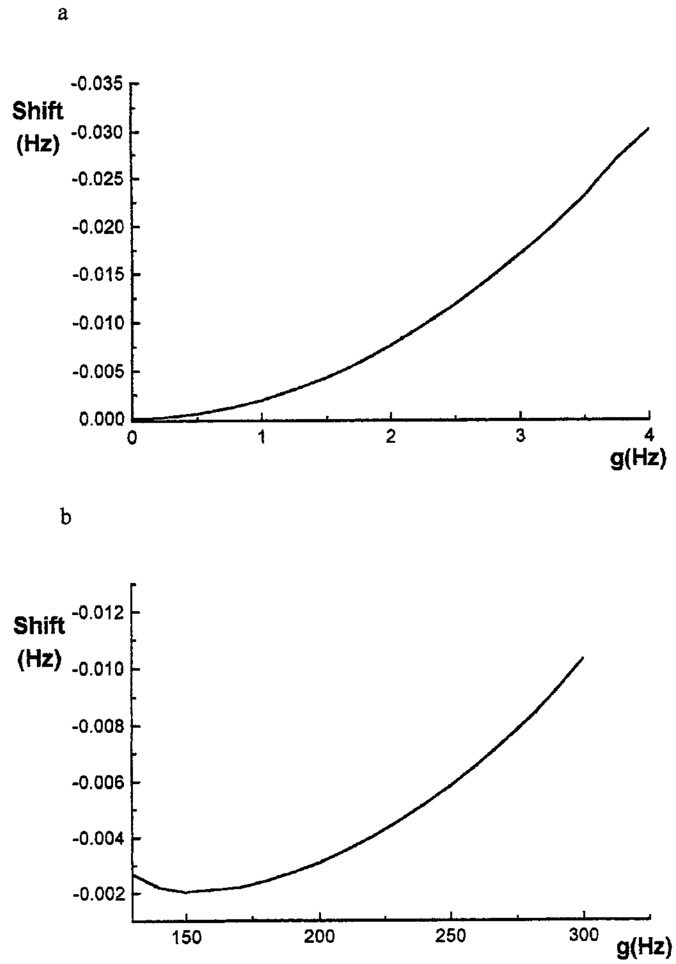

Fig. 5. The frequency shifts for the single-quantum (a) and four-quantum (b) resonances induced by driving field in units of Γ_0 (in Herz).

Acknowledgements

This work has been supported by International Science Foundation. E. Alexandrov is grateful also to Alexander von Humboldt Stiftung.

Note added in proof

The expected features of the 4-quantum resonance have been recently confirmed experimentally in collaboration with Dr J. L. Rasson (Institut Royal Meterologique de Belgique, Belgium), will be published in "Optics & spectroscopy" in 1997.

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