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Multiple-quantum radio-frequency spectroscopy of atoms: application to the metrology of geomagnetic fields

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The evolution of the radio-frequency magnetic resonance spectrum of optically oriented potassium atoms in terrestrial magnetic fields as a function of the amplitude of the alternating magnetic field H_1 is investigated theoretically and experimentally. It is shown that among the set of observed n-quantum resonances which satisfy the selection rules $\Delta F=0$ and $\Delta m_F=\pm n$ (n is an integer), there is one resonance with the highest multiplicity $n=4$ ($m_F=2\Leftrightarrow m_F=-2$), which has exceptional properties. These qualities make this resonance a candidate for use in a weak-field quantum magnetometer with record-high characteristics. A preliminary investigation of an experimental model of a four-quantum potassium magnetometer is performed. © *1999 American Institute of Physics.* [S1063-7842(99)00709-6]

INTRODUCTION

The Zeeman effect in the ground-state radio-frequency spectra of several alkali-metal atoms has been utilized since the late 1950s for highly accurate measurements of the absolute value of the induction of weak magnetic fields mainly in the geomagnetic range. The demands for precision measurements of this kind are very diverse, and the accuracy requirements imposed have no upper bound, creating a situation that constantly stimulates the search for new solutions. The greatest absolute accuracy and variational sensitivity have been achieved using an optically pumped potassium-vapor magnetometer, which utilizes a narrow isolated resonance with $\Delta F=0$ and $m_F=2\Leftrightarrow m_F=1$ in the ground-state radio-frequency spectrum of one of the two stable isotopes of potassium.[1] Apart from the one-quantum transitions with $\Delta m_F=\pm 1$, multiple-quantum resonances with $\Delta m_F=\pm n$ appear when the amplitude of the resonance-stimulating alternating magnetic field H_1 is increased, and they are very effective in the quasiequidistant magnetic splitting system of the ground-state hyperfine sublevels. At the time of their discovery in the early 1950s,[2,3] these resonances seemed attractive because of their intrinsic width, which successively decreases with increasing n. However, it was soon found that their frequency depends on the amplitude of the alternating field and that the stronger it is, the higher is the order of the resonance. This dramatically reduced the interest in these resonances, and since then they were not employed in magnetic-field metrology.

In this report we turn our attention to the special properties of the resonance with the highest multiplicity $n=4$, which is essentially not displaced by the alternating field. In addition, the frequency of this resonance (in contrast to all the others) exhibits a strict linear dependence on the induction of the static magnetic field. Being very narrow and very intense, this resonance merits study as a candidate for use in a quantum magnetometer with record-high characteristics.

The easily predicted features of the four-quantum resonance might have provided an exceptional experimental technique back in the 1950s. However, in the experiments at that time experimentalists focused on three-quantum resonances for technical reasons: the four-quantum resonance would have required a radio-frequency field with an excessively high amplitude. Under our conditions we worked with such narrow resonances and in such weak static fields that only negligible amplitudes were needed to excite the four-quantum resonance.

ANALYSIS OF THE EVOLUTION OF THE MAGNETIC RESONANCE SPECTRUM OF POTASSIUM

In a weak magnetic field the ground state of the potassium atom forms two systems of almost equidistant magnetic sublevels separated by a hyperfine interval. An example of the energy structure of the ^{39}K isotope is shown in Fig. 1a, where the magnetic splitting has been greatly magnified for clarity. In a terrestrial field the spectrum of transitions with $\Delta m_F=\pm 1$ consists of six closely arranged lines in the vicinity of a frequency equal to $H\cdot 7$ Hz/nT, where H is the induction of the field in nanoteslas. The exact values of the frequencies of these transitions obey the familiar Breit–Rabi formula and were given in Ref. 4. Under the conditions of optical orientation the transitions within the $F=2$ state are most clearly expressed in the spectrum, being manifested by four almost exactly equidistant lines separated by an interval equal to $2bH^2$, where the value of b for the ^{39}K isotope is 106.327 GHz/T^2. In an average terrestrial field of 50 μT this interval amounts to about 500 Hz.

In a vacuum flask with a paraffin cover the intrinsic width of the resonances of potassium vapor is of the order of 1 Hz. The possibility of using one completely isolated, very strong line in the spectrum gives potassium a decisive advantage over cesium, which has been most popular in quantum magnetometry, but whose numerous overlapping resonances form a broad (of the order of 50–100 Hz) asymmetric line with an indefinite position for the maximum, creating a ma-

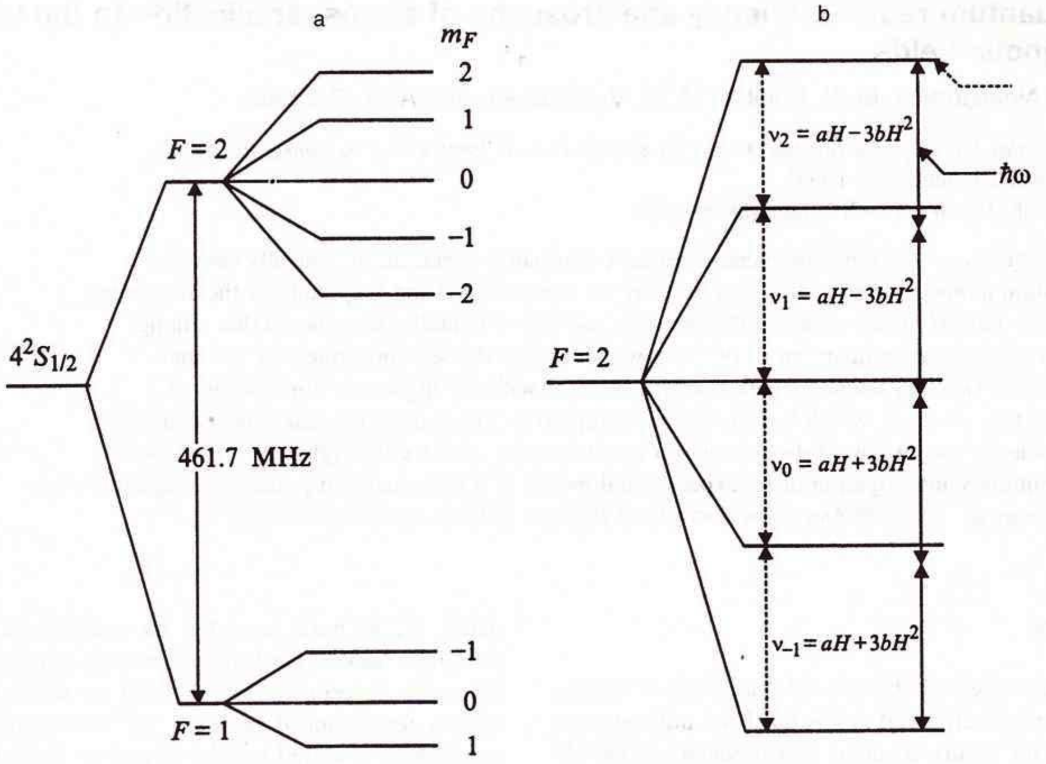

FIG. 1. Scheme of the ground-state energy sublevels of the ^{39}K atom (a) and detailed structure of the magnetic splitting of the upper ground-state hyperfine sublevel of the ^{39}K atom in a magnetic field with induction H with accuracy to the terms quadratic with respect to the field for $a=7.004666$ GHz/T and $b=106.327$ GHz/T^2 (b).

jor source of systematic errors in cesium magnetometers. Potassium magnetometers are completely free of errors associated with the influence of nearby resonances. The small width of the principal resonance together with its high amplitude (the signal-to-noise ratio is of the order of 10^4 in a 1-Hz band) ensures the record-high sensitivity of potassium magnetometers. Figure 2 presents a fragment of a record of the difference between the readings of two independent potassium magnetometers, whose sensors were separated by two meters. The differential record enabled us to suppress the natural fluctuations of the Earth's magnetic field by a factor of 100 and to approach the internal noise of the instruments. The noise track presented, which is confined to 1/4 pT, gives an upper estimate of the noise of the instruments. The record was made at the rate of 5 readings per second and provided a picture of the resolving power achieved in the instrument.

A spectrum of 6 isolated lines is observed for a fairly low amplitude of the radio-frequency field H_1, which is assigned by the relation $\gamma H_1 \leq \Gamma$, where γH_1 is the matrix element for a transition between adjacent sublevels and Γ is the intrinsic width of the transition. At such a low amplitude of H_1 each atom responds to the perturbation as a set of independent two-level systems. As H_1 is increased, the spectrum begins to change: the lines broaden and shift, and new lines corresponding (in terms of perturbation theory) to multiphoton resonances appear. Field-induced line broadening is well known in the theory of magnetic resonance. The field-induced displacement of a resonance in a two-level system is also well known as the Bloch–Siegert effect. However, a far stronger field-induced shift of the resonance is exhibited in multilevel quasiequidistant systems. In second-order perturbation theory the energy shift ΔE_j of a certain level j is given by the expression[5]

$$\Delta E_j = \Sigma_k |V_{jk}|^2/(E_j - E_k + \hbar\omega), \tag{1}$$

where $|V_{jk}|$ is the matrix element associating the jth level with any possible kth level having an energy E_k, and $\hbar\omega$ is the quantum energy of the field H_1.

The formula presumes that the kth level lies above the jth level (otherwise, the sign in front of $\hbar\omega$ changes), and the denominator is fairly large compared to the width of the transitions, i.e., the displacement of levels under the action of a nonresonant field is considered (a resonant field does not

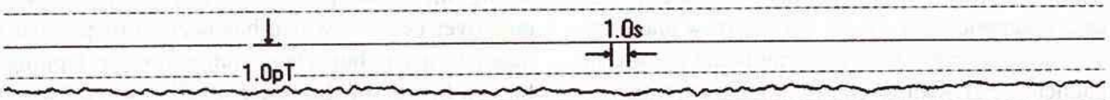

FIG. 2. Experimental record of the difference between the readings of two commercial GEM Systems potassium magnetometers in the Earth's natural field (Canada, April 1998).

shift the levels). The frequency shift of a transition is calculated as the difference between the energy shifts of the combining levels. As H_1 increases, the field-induced shift increases quadratically and becomes significant in the region where multiple-quantum resonances appear, increasing with increasing multiplicity of the resonance. The resonance of highest multiplicity is an exception. This can be seen by applying formula (1) to the calculation of the shifts of the $m_F=2$ and $m_F=-2$ levels under the action of the field H_1 with the frequency aH (Fig. 1b), which corresponds to 1/4 of the frequency of the unperturbed gap between the $m_F=2$ and $m_F=-2$ levels. This frequency exceeds the resonance value for the $m_F=2 \Leftrightarrow m_F=1$ transition by $3bH^2$, while it falls short of the resonance value for the $m_F=-1 \Leftrightarrow m_F=-2$ transition by the same amount. As a result, the upper and lower sublevels of the $m_F=2 \Leftrightarrow m_F=-2$ transition are shifted identically in the same direction, so that the transition frequency itself remains unchanged (Fig. 1b).

The conclusion that there is exact compensation of the field-induced shifts of the levels is based on the use of perturbation theory and requires more definitive confirmation for two reasons. First, the excitation of the four-quantum resonance near saturation requires a fairly high value for H_1, which violates the criterion for a small perturbation. Second, in arriving at the conclusion that the field-induced shifts are compensated, the lines of the one-quantum resonances were assumed to be strictly equidistant, i.e., the terms higher than quadratic in the expansion of the transition frequencies in powers of H were discarded. In addition, the influence of the wings of neighboring resonances that are broadened at high values of H_1 on the position of the maximum of the four-quantum resonance should also be taken into account. For all these reasons it was decided to carry out an exact solution of the problem of magnetic resonance in an 8-level system under the conditions of optical pumping without restrictions on the amplitude of the field H_1. For this purpose we solved the Liouville equation for the density matrix, which was supplemented by a phenomenological relaxation matrix describing the optical pumping. During the search for a stationary solution in the rotating-field approximation, a system of 34 differential equations was reduced to a system of algebraic equations, which was solved using a personal computer for a grid of values of the frequency and amplitude of H_1.

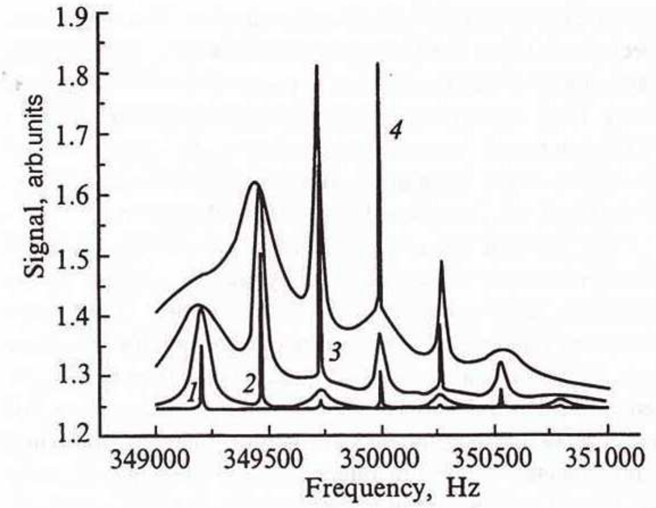

FIG. 3. Calculated ^{39}K magnetic resonance spectrum for four values of the field amplitude: *1)* 2π, *2)* $2\pi\cdot 20$, *3)* $2\pi\cdot 80$, *4)* $2\pi\cdot 190$ Hz. The calculation was performed for a static field H with an induction of about 50 μT.

The result is presented in Fig. 3 (Ref. 4). The lowest spectrum corresponds to the condition $\gamma H_1=2\pi$ Hz (the intrinsic width of the resonances was assumed to be equal to 1 Hz). The four resonances in the $F=2$ state can be seen here. The resonances in the $F=1$ state are too small for this scale. When γH_1 is increased to $2\pi\cdot 20$ Hz, the one-quantum resonances are strongly broadened and appreciably displaced, while the three two-quantum resonances are optimized. When $\gamma H_1=2\pi\cdot 80$ Hz, the two three-quantum resonances achieve maximum steepness, while the two-quantum resonances have already reached saturation and broadened. Finally, when $\gamma H_1=2\pi\cdot 190$ Hz, the four-quantum resonance dominates. An analysis showed that the frequency of this resonance responds to a 10% change in H_1 no more strongly than does the frequency of a one-quantum resonance in the case of the analogous change in H_1 in the vicinity of the optimum value $\gamma H_1=2\pi$ Hz, which is of the order of 50 fT and is thus essentially a negligible quantity. The steepness of the four-quantum resonance at the optimum is 7 times higher than the steepness of a one-quantum resonance.

Thus, the mathematical simulation confirmed the expected promising nature of the four-quantum resonance. This conclusion has also been confirmed by experimental testing.[6] In addition to the data presented in Ref. 6, we present a set of panoramic magnetic resonance spectra, which were recorded by the technique of low-frequency modulation of the frequency of H_1. The signal was recorded from the output of a synchronous detector, so that the resonance lines had the from of dispersion curves. In Fig. 4 the upper spectra correspond to higher intensities of H_1. The lowest spectrum displays the one-quantum resonances: three belong to the $F=2$ state and one of the $F=1$ resonances. In the next spectrum two two-quantum resonances dominate. The third spectrum displays broadened two-quantum resonances and

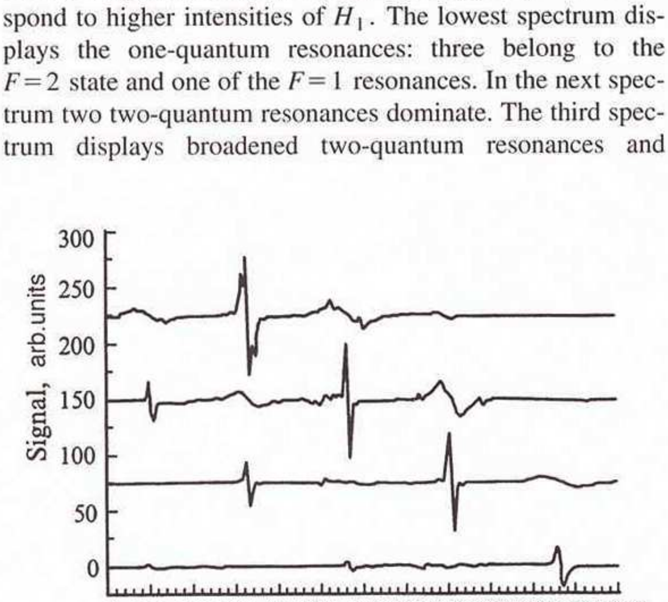

FIG. 4. Experimental potassium magnetic resonance spectra for various values of H_1. The spectra were recorded in a field with a magnetic flux density of about 50 μT. The frequency of the field H_1 measured relative to the doubled resonance frequency in cesium vapor, which was used to stabilize the magnetic field, is plotted along the horizontal axis.

clearly expressed three-quantum resonances. The uppermost spectrum exhibits the four-quantum resonance. The complicated shape of the resonances is associated with the excessively high modulation frequency superimposed on H_1, which generated nonstationary responses. We also note that the widths of the resonances are restricted by the instrumental width of the procedure for obtaining the spectra.

The use of the four-quantum resonance for the purposes of magnetometry is complicated by the narrowness of the resonance, which requires very slow scanning of the resonance and thus causes the instrument to have a low response rate. Therefore, a ''tandem'' scheme combining a self-generating cesium magnetometer, which provides for a fast response of the instrument, and a four-quantum potassium magnetometer, which guarantees a high absolute accuracy and linearity of the output frequency with respect to the measured field, was chosen. An instrument of this type is presently undergoing laboratory testing. Its resolving power is determined by the cesium magnetometer and is of the order of 1 pT/Hz$^{1/2}$ (the rms noise). The long-term stability will apparently be limited by the photoinduced shifts of the potassium $m_F=2$ and $m_F=-2$ levels, which can be minimized by lowering the intensity of the optical pumping of potassium, if we bear in mind the high steepness of the four-quantum resonance.

CONCLUSION

The potassium atom in a fairly strong alternating field represents a new quantum system, which has, in particular, a new magnetic resonance line that is not displayed by the free atom. Until now, only one electronic paramagnet with a linear magnetic-field dependence of the resonance frequency, viz., ^{4}He in the metastable 2^3S_1 state, has been available to quantum magnetometry. The four-quantum potassium resonance has the same property, but is also a hundred times narrower. This circumstance allows us to recommend this resonance for standardizing the induction of fields in the geomagnetic range.

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