Adiabatic Transfer between Hyperfine Levels in Combined Electric and Magnetic Fields

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We report observation of transfer between hyperfine levels due to the action of combined external electric and magnetic fields, changing the energy and the angular momentum of the atom. The experiments were performed on a beam of Ga atoms in the fine structure level $4s^2 4p\,^2P_{3/2}$, traversing specially confined and spatially displaced perpendicularly crossed electric and magnetic fields. The hyperfine level transfer was detected by probing the population of the levels involved. [S0031-9007(96)01184-2]

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In atomic physics as well as in quantum optics transfer of population between atomic or molecular energy levels is an important and necessary tool to prepare well defined initial conditions for experimental investigations. Different methods have been developed and sucessfully used: application of π pulses [1], adiabatic transfer [2,3], counterintuitive pulse sequences [4–7], and adiabatic passage in a light field [8–10]. Most related to this work is adiabatic transfer through level anticrossings. It was pointed out by Weider and Eck [11] in 1967 that crossings of hyperfine sublevels in magnetic fields can be converted to anticrossings by introducing an electric field perpendicular to the magnetic field. This fact was extensively used in level anticrossing spectroscopy [12], e.g., intensity changes due to electric field induced anticrossings have been observed by Eck and Huff [13] and by the group of Kleinpoppen [14] in ionized helium and by Adler and Malka [15] in Li I. Rubbmark *et al.* [3] have used pure electric field induced level anticrossings to lower the ionization limit of certain sublevels of *n*-manifolds of Rydberg states in Li I. The aim of this work is to report on direct experimental observation of a new type of adiabatic transfer in quasistatic combined external electric and magnetic fields working without application of electromagnetic radiation.

An atom in the initial level F_i experiences first a magnetic field, then additionally an electric field oriented perpendicularly to the magnetic one, and later on only the electric field before entering a field free region. The increasing and decreasing of the fields must be slow enough to ensure abiabatic energy changes. After this field cycle the atom can be found in another final level F_f. The atom changes its hyperfine energy and its angular momentum without interaction with electromagnetic radiation. The adiabatic transfer from initial to final state takes place while the atom passes through successive level crossings and anticrossings of the atomic levels modified by the applied electric and magnetic fields.

This result of adiabatic transfer was found during extensive theoretical and experimental investigations of the sodium D_2 line ($3\,^2S_{1/2}$-$3\,^2P_{3/2}$) under the influence of the simultaneous action of external electric and magnetic fields, with either parallel, perpendicular, or arbitrary relative directions [16]. The influence of the above mentioned field cycle on the hyperfine levels of Na $3\,^2P_{3/2}$ was discussed theoretically in [17]. To ensure the appearance of level crossings between hyperfine states with $\Delta M_F = \pm 2$ in magnetic fields and corresponding anticrossings in perpendicularly crossed electric and magnetic fields the atom must have a tensor polarizability $\alpha_2 \neq 0$ (and therefore $J > 1/2$) and a nuclear spin quantum number $I \geq 1/2$.

Figure 1 shows the influence of the fields on the hyperfine levels of $4s^2 4p\,^2P_{3/2}$ for the isotope ^{69}Ga (for the choice of Ga see below). In the first section ($0 \rightarrow B_{\max}$,

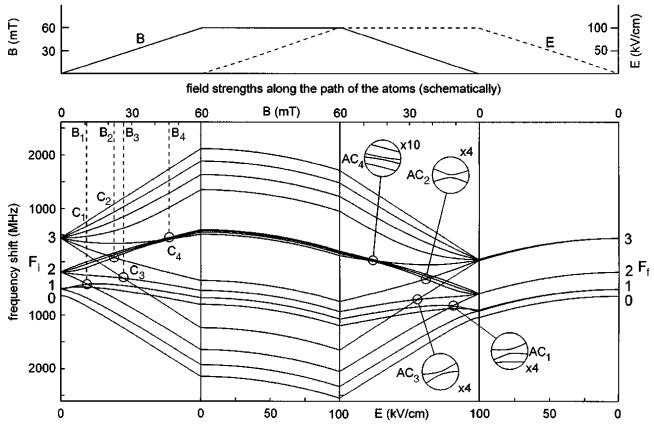

FIG. 1. Behavior of the hyperfine states in the applied field cycle for ^{69}Ga.

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$E = 0$) the magnetic field leads to four $\Delta M_F = \pm 2$ crossings $(C_1, \ldots, C_4)$ appearing at magnetic field strengths ${}^{69}B_1, \ldots, {}^{69}B_4$. In the next section ($B = B_{\max}, 0 \rightarrow E_{\max}$) the electric field shifts all hyperfine states nearly parallel. Then the magnetic field decreases to zero whereas the electric field is further acting ($B \rightarrow 0, E = E_{\max}$). The electric field, oriented perpendicularly with respect to the B field, converts the crossings into anticrossings $(AC_1, \ldots, AC_4)$. In the last section the electric field is switched off ($B = 0, E \rightarrow 0$). Across the region of the avoided crossings the character of the state changes from F_i to F_f. This applies to all levels except $F_i = 0$, which experiences no crossing or anticrossing and therefore always leads to $F_f = F_i = 0$.

In the theoretical analysis of the dependence of the energy eigenvalues $\mathcal{E}_{F,M_F}(B, E)$ on the magnetic and electric field strengths we considered energy surfaces above the (B, E) plane. The four sections of Fig. 1 represent cuts through those energy surfaces parallel to the $(\mathcal{E}, E)$ and $(\mathcal{E}, B)$ planes, respectively. For perpendicular fields the atom-field system retains just one symmetry. This entails that the system of levels decomposes into two noninteracting systems corresponding to odd or even M_F. So there are also two uncoupled systems of energy surfaces. Some surfaces have a common point for certain values of B and E corresponding to a crossing. There are four (three) such points with $E = 0$ in the odd (even) system besides some other points with $E \neq 0, B \neq 0$. Near such a point the upper (lower) energy surfaces develop a downward (upward) groove, whose walls are nearly straight in the E direction, reflecting the linear dependence of the Hamiltonian on B. The crest and bottom lines of the grooves have a parabolic shape with a common tangent in the point where the surfaces touch; this results from the E^2 dependence of the effective electric field Hamiltonian. When a level crossing in a field cycle occurs (first section of Fig. 1) the system passes from one surface onto the other through the common point while $E = 0$; thereafter it is confined to this surface when $E \neq 0$ (third section of Fig. 1).

In the experimental realization one needs to consider carefully how to implement the above mentioned field cycle. In a "gedanken experiment" the experiment may be performed on an atom at rest and with an arbitrarily slow rate of field changes. In reality, we replace the time varying fields, acting on an atom at rest, by a linear arrangement of spatially confined static fields, produced by a special field arrangement (total length ca. 8 cm). Atoms of an effusive beam traverse the fields. In their own frame of reference the atoms experience time dependent fields. Some of the atoms change their hyperfine level from F_i to F_f during passage through the fields.

Prerequisite to the detection of population transfer between initial F_i and final F_f hyperfine levels is the establishment of an imbalance in the state population between those levels. In our experiment this is done by a pump-probe technique. A sketch of the experimental arrangement is shown in Fig. 2(a). In Fig. 2(b) the principle of the pump and probe experiment is shown. We optically pump all atoms from level F_f to other hyperfine or fine structure levels before the atoms enter the field arrangement. The linearly polarized probe laser light (parallel to B, direction y in Fig. 2), acting on the atoms which have left the field arrangement, excites the same transition as the pump laser. If the combined action of both fields causes a transfer from level F_i to level F_f, fluorescence is observed.

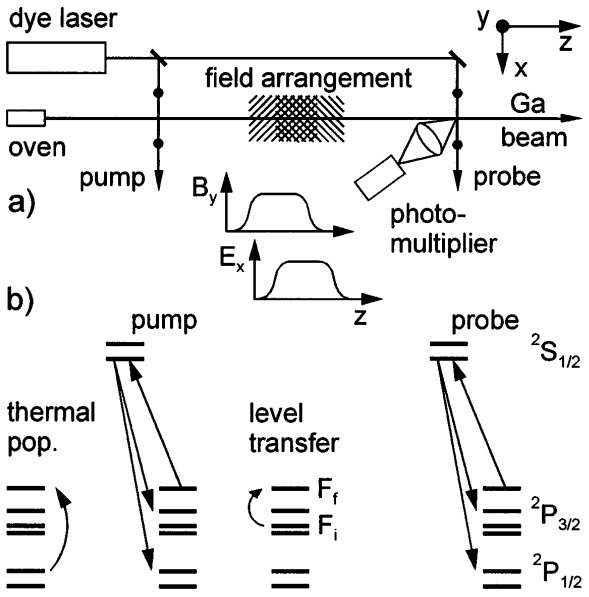

FIG. 2. (a) Sketch of the experimental arrangement; (b) scheme of the pump-probe detection. For details see text.

Of course the atoms have to stay in the same fine structure level during the time of flight through the field arrangement. To experimentally demonstrate the process we need to find atoms which have a suitable ground or long-living metastable levels. One of the elements which can fulfill these criteria is gallium. The metastable level $4s^2 4p\,{}^2P_{3/2}$, suitable for our investigations, has an excitation energy corresponding to 826 cm^{-1} and is sufficiently populated by thermal collisions in the oven ($T = 1670$ K) used for producing the atomic beam. The population can be monitored by exciting the transition $4s^2 4p\,{}^2P_{3/2} \rightarrow 4s^2 5s\,{}^2S_{1/2}$, $\lambda = 417.206$ nm. Ga in its natural abundance has two isotopes, ^{69}Ga (60%) and ^{71}Ga (40%), both having the same nuclear spin quantum number, $I = 3/2$. Therefore the hyperfine structure of the lines of both isotopes is comparable to the sodium D_2 line concerning the number and relative intensity of the components. The hyperfine constants of ^{69}Ga in its ${}^2P_{3/2}$ level are $A = 190.794(5)$ MHz, $B = 62.522(12)$ MHz, whereas those of ^{71}Ga are $A = 242.434(5)$ MHz, $B = 39.399(10)$ MHz [18]. The hyperfine splitting of ^{71}Ga is therefore larger, and consequently the values ${}^{71}B_1, \ldots, {}^{71}B_4$ (16.1; 28.8; 33.5; 57.7 mT) occur at higher field strengths compared to the values ${}^{69}B_1, \ldots, {}^{69}B_4$ (11.2; 23.0; 27.0; 45.3 mT) in Fig. 1 [calculated using

$\alpha_0 = 0.08$ MHz/(kV/cm)^2; $\alpha_2 = -0.00276$ MHz/(kV/cm)^2; $g_I' = -7.239$] [19]. We will refer in particular to this point when discussing the observed spectra.

Figure 3 shows the fluorescence spectra obtained exciting the hyperfine components of the $^2P_{3/2} \rightarrow {}^2S_{1/2}$ transition subsequently while scanning the probe laser frequency. The intensity scale is the same for all spectra. Spectrum 3(a) was obtained while blocking the pump beam and switching off the electric and magnetic field. The hyperfine components are labeled as $^{69}T_{F_f,F'}$ or $^{71}T_{F_f,F'}$, to identify the isotope and the quantum numbers

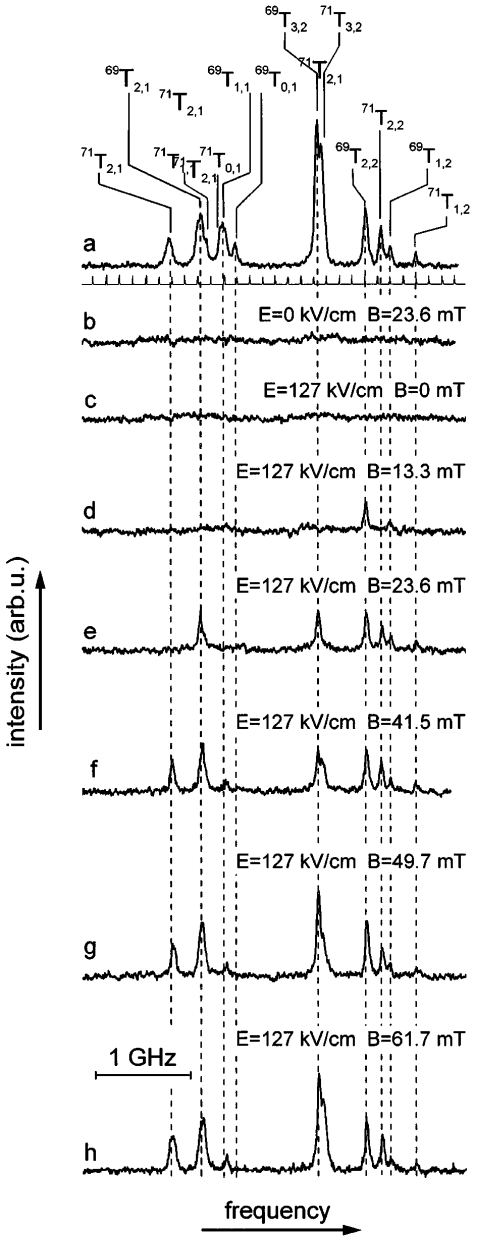

FIG. 3. Fluorescence spectra induced by the probe laser. (a) $E = B = 0$, the pump beam is blocked. All hyperfine components of both isotopes are resolved. Below the spectrum, the trace of the marker etalon is shown. (b)–(h) Spectra with activated pump beam for different field strength combinations. For an explanation, see text.

F_f of the lower and F' of the upper hyperfine level. In the other spectra of Fig. 3 the pump beam was activated. Figures 3(b) and 3(c) show spectra where either only the magnetic or only the electric field was switched on to the values indicated in the figures. No fluorescence is observed in the probe region. This proves that the pump beam depopulates the levels completely.

In all other spectra the electric field had the strength of 127 kV/cm. In spectrum 3(d) the magnetic field strength is chosen above the value $^{69}B_1$ for the first crossing and anticrossing C_1/AC_1 (coupling $F = 1$, $M_F = 0 \leftrightarrow F = 2$, $M_F = -2$) in ^{69}Ga, but below $^{71}B_1$. Figure 1 shows that for magnetic field strengths below B_1 no level transfer can occur. Therefore we observe in this spectrum only two components, due to C_1/AC_1: $^{69}T_{2,2}$ and $^{69}T_{1,2}$. Because the pump and the probe laser beams were linearly polarized (parallel to $\vec{B}$), only transitions $\Delta M_F = 0$ are excited. As seen in Fig. 1 and detailed in Ref. [17], we repopulate the states $F_f = 2$, $M_F = -2$, and $F_f = 1$, $M_F = 0$ due to C_1/AC_1. From these states, π transitions to the hyperfine states belonging to $F' = 2$ can be excited, but not to states belonging to $F' = 1$. Indeed, these last components are missing in spectrum 3(d). In spectrum 3(e) the field strength is chosen above $^{69}B_2$ but between $^{71}B_1$ and $^{71}B_2$. For ^{69}Ga now C_2/AC_2 (coupling $F = 3$, $M_F = -2 \leftrightarrow F = 2$, $M_F = 0$) also occurs, while for ^{71}Ga only C_1/AC_1 is possible. Therefore we find in the spectrum apart from the components $^{69}T_{2,2}$, $^{69}T_{1,2}$, the new components $^{69}T_{2,1}$, $^{69}T_{3,2}$, $^{71}T_{2,2}$, and $^{71}T_{1,2}$. In spectrum 3(f) the magnetic field is above $^{69}B_3$ and $^{71}B_3$. Compared to spectrum 3(e) we now observe in addition the components $^{71}T_{2,1}$ and $^{71}T_{3,2}$. Component $^{69}T_{2,1}$ has gained more intensity since $^{69}F_f = 2$ regains population because of the two crossings and anticrossings C_2/AC_2 and C_3/AC_3 (coupling $F = 3$, $M_F = -3 \leftrightarrow F = 2$, $M_F = -1$). The increase of $^{69}T_{2,2}$ is less pronounced since the transition probability for the π excitation to $F' = 2$ is lower for $M_F = -1$ than for $M_F = -2$. Because of imperfections of the experiment (increasing stray magnetic field in the probe region) we also observe the components $^{69}T_{1,1}$ and $^{71}T_{1,1}$ with small intensity. In spectrum 3(g) the magnetic field is strong enough to exceed $^{69}B_4$, but not $^{71}B_4$, and component $^{69}T_{3,2}$, in particular, gains more intensity from the crossing and anticrossing C_4/AC_4 (coupling $F = 3$, $M_F = -1 \leftrightarrow F = 2$, $M_F = 1$). This explains why the intensity ratio $^{69}T_{3,2}/^{71}T_{3,2}$ is not determined by the relative isotope abundance [cf. Fig. 3(a)]. In the last spectrum, Fig. 3(h), the field strength is high enough to exceed B_4 for both isotopes, and the intensity ratio $^{69}T_{3,2}/^{71}T_{3,2}$ is now the same as in Fig. 3(a). What is missing in Fig. 3(h) compared to Fig. 3(a) are the components $^{69}T_{0,1}$ and $^{71}T_{0,1}$, for which no hyperfine level transfer and therefore no repopulation is possible.

The transfer efficiency seems to be rather high when estimating from the intensity of the spectra in Fig. 3. However, the population monitored by the probe laser is gained not only from the original population of the levels but also

from optical pumping in the first (pump) interaction region. As can be derived from Fig. 1, only special F, M_F states are undergoing the crossing-anticrossing pairs. For example, if all population is pumped to $F_i = 1, M_F = 0$, one will find after the field cycle all population in state $F_f = 2, M_F = -2$. In this case the efficiency will be unity. If all population is pumped to $F_i = 1$, the efficiency is $1/3$, because the states $M_F = \pm 1$ will not be transferred to $F = 2$. Besides, the transfer efficiency is dependent on how well the atom follows the adiabatic curve through the region of the anticrossings. If the energy spacings are too small or if the atomic velocity is too high, the atom may undergo diabatic transitions described by the Landau-Zener formula [3,20–22]. This is the reason why the electric field has to be sufficiently strong to ensure the energy gap to be large enough and furthermore to ensure that most of the atoms in the thermal beam follow the curves adiabatically. The adiabacity condition was estimated to be $\Delta\omega\tau \gg 1$, where $\Delta\omega = \Delta E/\hbar$ measures the energy gap at the anticrossing point and τ is the time for passing the anticrossing region. From the B-field gradient in our field arrangement we get the conditions fulfilled for atoms having velocities $v \ll 5 \times 10^3$ m/s, a value large compared to the thermal mean velocity in the beam (630 m/s, $\tau \approx 0.5$ μs, $\Delta\omega\tau \approx 10^{-2}$).

In conclusion, we have shown that atoms in a beam, crossing spatially confined perpendicularly crossed electric and magnetic fields, undergo transitions between hyperfine levels. The comparison of the results from the theoretical model and from the experiment allowed us to unambiguously identify the states involved in the transfer. This method should be applicable for all atoms having in their ground levels $J > 1/2$ and $I \geq 1/2$ and also for a large variety of molecules.

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