Strongest Magnetically Induced Transitions
in Alkali Metal Atoms

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Atomic transitions in alkali metals that have zero probability in the absence of a magnetic field but have large probabilities in the presence of a magnetic field are called magnetically induced (MI). They are of interest because of their large probabilities, which exceed the probabilities of usual transitions in a wide magnetic field range. Magnetically induced transitions are classified as type-1 (MI1) and type-2 (MI2) and their total number is about 100. In this work, MI2 transitions are examined between ground $F_g$ and excited levels $F_e$ of the hyperfine structure satisfying the condition $F_g - F_e = \Delta F = \pm 2$, which are forbidden in zero magnetic field but have large probabilities in the presence of a magnetic field. The probabilities of the MI2 transitions with $\Delta F = +2$ and the MI1 transitions with $\Delta F = -2$ are maximal in the case of optical radiation with the $\sigma^+$ and $\sigma^-$ polarizations, respectively. This difference is called type-1 magnetically induced circular dichroism (MICD1). It has been shown for the first time that the probability of the strongest MI2 transition in the $^{85}$Rb atom corresponding to the $D_2$ line in magnetic fields $> 100$ G in the case of $\sigma^+$ radiation is larger than the probability of the strongest MI2 transition in the case of $\sigma^-$ radiation by a factor of 2.5. This difference is called type-2 magnetically induced circular dichroism (MICD2). It has been shown how to determine the strongest MI transition for any alkali metal atom, which is important for its application in magneto-optical processes. Theoretical curves reproduce well experimental results.

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It is known that the probability (intensity) of atomic transitions in alkali metals can significantly change in high magnetic fields [1, 2]. A bright example of a giant increase in the probability is the behavior of magnetically induced (MI) transitions in Cs, Rb, K, etc., atoms, which constitute a large class consisting of about 100 atomic transitions with interesting important features [3–10]. These transitions in zero magnetic field are forbidden by selection rules, whereas their probabilities increase significantly in an external magnetic field; for this reason, they are called MI transitions. Significant change in the probabilities of transitions, in particular, a giant increase in the probabilities of MI transitions, is due to the effect of “mixing” of magnetic sublevels for ground $F_g$ or excited levels $F_e$ with magnetic sublevels of the nearest transition; the mixing effect is induced by the external magnetic field [3, 4, 7–11]. Magnetically induced transitions are of interest because their probabilities in certain wide ranges of magnetic fields can be much larger than the probabilities of ordinary atomic transitions allowed in zero magnetic field. The strength of interaction of an atom with the magnetic field can be quantitatively characterized by the characteristic magnetic field $B_0 = A_{\text{hlb}}/\mu_B$, where $A_{\text{hlb}}$ is the magnetic dipole constant of the ground level of the atom and $\mu_B$ is the Bohr magneton [12, 13]. For $^{85}$Rb and $^{87}$Rb atoms, $B_0 = 0.7$ and 2.4 kG, respectively.

Magnetically induced transitions are separated into type-1 (MI1) and type-2 (MI2) MI transitions [14]. In terms of the representation $|F, m_F\rangle$, where $F$ is the total angular momentum of the atom and $m_F$ is its projection, MI1 transitions are transitions $|F_g, 0\rangle \rightarrow |F_g, F_e\rangle$ between the ground $F_g$ and excited levels $F_e$ (the prime marks upper excited levels) with zero probability in zero magnetic field. The probability of these transitions increases abruptly with the applied magnetic field and asymptotically approaches a constant value with a further increase in the magnetic field $B \gg B_0$ [4].

Type-2 magnetically induced transitions are transitions $|F_g, m_F\rangle \rightarrow |F_e, m_F'\rangle$ between the ground $F_g$ and excited levels $F_e$, where $F_e = F_g \pm 2$ and $m_F' - m_F = 0, \pm 1$. The probability of MI2 transitions increases abruptly with the applied magnetic field, but the probability of these transitions again tends to zero with a further increase in the magnetic field $B \gg B_0$. In this
work, we consider M12 transitions corresponding to the $^{85}$Rb $D_2$ atomic line. In [8–10], we revealed the following property of the dependence of the intensity of MI transitions on the polarization of exciting radiation: the intensities of atomic transitions with $F_e - F_g = \Delta F = +2$ are maximal in the case of exciting radiation with the circular polarization $\sigma^+$ when $m_{F_e} - m_{F_g} = +1$, whereas the intensities of atomic transitions with $F_e - F_g = \Delta F = -2$ are maximal in the case of exciting radiation with the circular polarization $\sigma^-$ when $m_{F_e} - m_{F_g} = -1$. The difference between the intensities of some MI transitions excited by $\sigma^+$ and $\sigma^-$ radiation can be large [9]. The difference in the response of an atomic system for absorption, fluorescence, resonant ionization of atoms, etc., in a nonzero magnetic field under excitation by $\sigma^+$ and $\sigma^-$ radiation is referred to in atomic spectroscopy as magnetically induced circular dichroism (MICD) [8, 9, 15]; the mentioned dichroism is called type-1 magnetically induced circular dichroism (MICD1).

In this work, we study type-2 magnetically induced circular dichroism (MICD2), the essence of which is as follows. Comparison of the probability of the strongest MI2 transition under the condition $F_e - F_g = \Delta F = +2$ induced by $\sigma^+$ radiation ($|2, -2 \rangle \rightarrow |4', -1' \rangle$ transition in $^{85}$Rb) with the probability of the strongest MI transition induced by $\sigma^-$ radiation ($|3, 0 \rangle \rightarrow |1', -1' \rangle$ transition in $^{85}$Rb) in a wide magnetic field range showed that the former probability is always larger. We previously showed [9] that the probability of the $2 \rightarrow 4'$ MI2 transition induced by $\sigma^-$ radiation (transition marked by red digit 5 in red square) is one-fourth of the probability of the transition marked by red digit 5 in red square. In other words, the probability of the strongest MI2 transition induced by $\sigma^+$ radiation is always larger than the probability of the MI2 transition induced by $\sigma^-$ radiation. In [10], we showed that the probability of the strongest $|3, -3 \rangle \rightarrow |5', -2' \rangle$ MI transition in the Cs atom corresponding to the $D_2$ line induced by $\sigma^+$ radiation in a wide magnetic field range is twice as large as the probability of the strongest $|4, -1 \rangle \rightarrow |2', -2' \rangle$ MI1 transition induced by $\sigma^-$ radiation.

Theoretical calculations show that the probability of the strongest MI2 transition induced by $\sigma^+$ radiation in all alkali metal atoms is always larger than the probability of the MI1 transition induced by $\sigma^-$ radiation. In particular, the probability of the strongest $|1, -1 \rangle \rightarrow |3', 0' \rangle$ MI2 transition that is induced by $\sigma^+$ radiation and corresponds to the $D_2$ lines of the $^{87}$Rb, $^{39}$K, and Na atoms is larger than the probability of the strongest $|2, +1 \rangle \rightarrow |0', 0' \rangle$ MI2 transition induced by $\sigma^-$ radiation by a factor of 4. Thus, the strongest MI2 transition is induced by $\sigma^+$ radiation, occurs with $\Delta F = +2$, and involves the magnetic sublevel with the minimum $m_F$ value for the ground level $F_e$.

Figure 1a shows the diagram of levels of $^{85}$Rb involved in the $D_2$ line and transitions induced by $\sigma^+$ radiation, $2 \rightarrow 4'$ MI2 transitions are marked by red circles; transition no. 5 in circle has the largest probability. (b) Shown are $3 \rightarrow 1', 2', 3', 4'$ transitions; $3 \rightarrow 1'$ MI2 transitions with small probabilities are marked by black circles; the $3 \rightarrow 4'$ transition marked as $^{85}$GT is called guiding atomic transition (GT).

Other important features of GT transitions are presented in [11]. Figure 2a shows the diagram of levels and $2 \rightarrow 1'$, $2'$, $3'$, $4'$ transitions induced by $\sigma^-$ radiation; $1'$ in square marks the only $2 \rightarrow 4'$ MI2 transition. Figure 2b shows the $3 \rightarrow 1'$, $2'$, $3'$, $4'$ transitions;
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MI\(_2\) transitions are marked by 1–, 2–, and 3– in red circles; 1– in red circle indicates the MI\(_2\) transition having the largest probability among them; the 3 → 4′ MI\(_2\) transition marked by 1′ in circle has the largest probability, and the 3 → 4′ transition marked as 85GT is called guiding atomic transition (GT).

MI\(_2\) transitions are marked by 1–, 2–, and 3– in red circles; 1– in red circle indicates the MI\(_2\) transition having the largest probability among them; the 3 → 4′ transition marked as 85GT is the guiding transition (GT). Below, the strongest MI\(_2\) transition induced by radiation and is marked by digit 5 in red circle is compared to the strongest transition induced by \(\sigma^-\) radiation and is indicated by 1− in red circle, and it is shown that transition marked by digit 5 in red circle is the strongest MI\(_2\) transition of the \(D_2\) line. Figure 3 shows the layout of the experimental setup. Radiation of a tunable diode laser with an external cavity [16] with a wavelength of 780 nm and a spectral width of about 1 MHz was used. To detect the absorption spectrum, we used a nanocell filled with Rb atomic vapor with the thickness in the direction of laser radiation equal to half the wavelength (\(L = \lambda/2 = 390 \text{ nm}\)) of radiation resonant with the \(D_2\) line. The nanocell was used to implement the \(\lambda/2\) method, which ensured the narrowing of atomic transitions (lines) in the absorption spectrum \(A(\nu)\) of the nanocell. To further narrow atomic lines, we performed double differentiation of the absorption spectrum \(A'(\nu)\), which ensured the additional significant narrowing of atomic lines in the second derivative (SD) of the spectrum [17]. This is particularly important for the frequency separation of closely spaced atomic transitions in the case of their large number. The nanocell was placed in a furnace with a hole for the passage of laser radiation and was heated to 110 °C, which ensured the atomic concentration \(N \approx 10^{13} \text{ cm}^{-3}\) (details of the design of the nanocell are presented in [18]). The nanocell was placed between strong permanent magnets (PM), which produced strong longitudinal magnetic fields, and the wave vector of laser radiation \(k\) was directed along the magnetic field \(B\) [19]. To form a frequency reference, a fraction of laser radiation was guided to a unit containing the additional nanocell (5) with the thickness \(L = \lambda/2 = 390 \text{ nm}\); the SD of the absorption spectrum of this nanocell was used as the frequency reference [17]. Optical radiation was detected by FD-24K photodiodes (4), signals from which were fed to a Tektronix TDS2014B oscilloscope (OS).

Upper lines Abs. in Fig. 4 are the experimental absorption spectra of the \(2, 3 \rightarrow 3', 4'\) transitions in the longitudinal magnetic fields \(B = (a) 800, (b) 900,\) and (c) 1000 G obtained by the \(\lambda/2\) method (\(L = \lambda/2 = 390 \text{ nm}\)) under irradiation by \(\sigma^-\) radiation (transitions are shifted from the frequencies of initial transitions at \(B = 0\) toward high frequencies). The power of the laser was 50 μW. As seen, some transitions in the absorption spectrum partially overlap. Red lines are SDs of absorption spectra (here and below, the SD is inverted for convenience). The \(2, -2 \rightarrow 4', -1'\) MI\(_2\)
transition marked by digit 5 in red circle is the strongest among the MI2 transitions in the $^{85}\text{Rb}$ atom induced by $\sigma^+$ radiation. The spectrum also includes the guiding transition $\text{GT} (^{85}\text{Rb})^+$ whose application is discussed below. Blue lines are the SDs of calculated absorption spectra for atomic transitions with a FWHM of 40 MHz. The calculations were performed within the theoretical model presented in [3, 4, 7, 8], which describes change in the probabilities and frequencies of atomic transitions in the magnetic field using the Hamiltonian matrix including all transitions inside the hyperfine structure. Lower lines Reper in Fig. 4 are the SDs of absorption spectra for $2 \rightarrow 1'$, $2'$, $3'$ transitions in $^{85}\text{Rb}$ and $1 \rightarrow 0'$, $1'$, $2'$ transitions in $^{87}\text{Rb}$ in zero magnetic field.

Since the aim was to compare the probability of the strongest MI2 transition with $\Delta F = +2$ in Rb induced by $\sigma^-$ radiation (transition marked by digit 5 in red circle) in the magnetic field with the probability of the strongest MI2 transition with $\Delta F = -2$ in Rb induced by $\sigma^-$ radiation (transition marked by $1^-$ in red circle), we present spectra under excitation by radiation with the $\sigma^-$ polarization. Upper lines Abs. in Fig. 5 are the experimental absorption spectra of the $3 \rightarrow 1'$, $2'$, $3'$, $4'$ transitions in $^{85}\text{Rb}$ induced by $\sigma^-$ radiation in the magnetic fields $B = (a)$ 800, (b) 900, and (c) 1000 G obtained by the $\lambda/2$ method ($L = \lambda/2 = 390$ nm). The spectra are at low frequencies and also include transitions corresponding to the $^{87}\text{Rb} D_2$ line, which are marked by arrows (only transitions important in this work are enumerated). We note that the $2 \rightarrow 4'$ MI2 transition induced by $\sigma^-$ radiation (transition marked in Fig. 2a by $1^-$ in square) is absent in the spectrum because it is shifted by $-8$ GHz and its amplitude is one-fourth of the amplitude of the MI2 transition marked by digit 5 in red circle. As seen, some transitions are poorly resolved in the absorption spectrum, but they are completely resolved in the SD of the absorption spectrum. Red lines are the SDs of the absorption spectrum of these atomic transitions. The $[3, 0] \rightarrow [1', -1']$ MI2 transition marked by $1^-$ in red circle is the strongest among the MI2 transitions in the $^{85}\text{Rb}$ atom induced by $\sigma^-$ radiation. The guiding transitions marked as $\text{GT} (^{85}\text{Rb})^-$ and $\text{GT} (^{87}\text{Rb})^-$ are also present in the spectrum. Blue lines are the SDs of calculated absorption spectra at a spectral width of transitions of 40 MHz. Lower lines Reper in Fig. 5 are the SDs of the absorption spectra of the $2 \rightarrow 1'$, $2'$, $3'$ transitions in $^{87}\text{Rb}$ in zero magnetic field. It is known that the parameters of cw diode lasers such as the power, spectral linewidth, and linearity of frequency scanning can depend on the operating frequency range of the laser. Since the MI2 transition indicated by digit 5 in red circle has a high frequency, whereas the MI2 transition marked by $1^-$ in red circle has a low frequency.

Fig. 4. (Color online) $^{85}\text{Rb}$ atom excited by $\sigma^+$ radiation; the thickness of the nanocell is $L = 390$ nm. Upper lines Abs. are the experimental absorption spectra of the $2 \rightarrow 3'$, $4'$ transitions in the magnetic fields $B = (a)$ 800, (b) 900, and (c) 1000 G. Red lines are the SD spectra, red digit 5 in circle marks the strongest of the indicated MI2 transitions, $\text{GT} (^{85}\text{Rb})^+$ is the guiding transition, blue lines are the calculated SD absorption spectra, and the FWHM is 40 MHz. Lower lines Reper are the SD absorption spectra of $^{85}\text{Rb}$, $2 \rightarrow 0'$, $1'$, $2'$ and $^{87}\text{Rb}$, $1 \rightarrow 0'$, $1'$, $2'$ transitions in zero magnetic field.
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The frequency distance between them is about 9 GHz at the magnetic field $B \approx 1000$ G, the direct comparison of the amplitudes of these transitions can be inappropriate. For this reason, we performed the following procedure. The probabilities of guiding transitions $GT$ ($^{85}\text{Rb}$)$^+$ and $GT$ ($^{85}\text{Rb}$)$^-$ are the same at any magnetic field [11]. The magnitude of absorption in the method is small, about 1%. Consequently, the magnitude of absorption can be written as $\sigma = A N L$, where $\sigma$ is the resonant absorption cross section, which is proportional to the probability of the atomic transition (depends on the magnetic field $B$ in our case); $N$ is the density of atoms; and $L$ is the thickness of the nanocell. Therefore, the amplitudes of transitions in absorption spectra are proportional to probabilities of these transitions (it is assumed that exciting radiation has a low intensity and does not saturate the transitions [20]). The MI2 transition marked by digit 5 in red circle is close in frequency to $GT$ ($^{85}\text{Rb}$)$^+$, and the MI2 transition indicated by $^-$ in red circle is close to $GT$ ($^{85}\text{Rb}$)$^-$. Consequently, measuring the ratio of the amplitudes of these MI2 transitions to the corresponding amplitudes of $GT$ ($^{85}\text{Rb}$)$^+$ and $GT$ ($^{85}\text{Rb}$)$^-$, one can determine the ratio $A_{\text{M12}}/A_{\text{M13}}$. In Fig. 6, filled squares are experimental results at the magnetic fields $B = 800, 900, 1000$ G, and the solid line is the theoretical curve. The reason for the choice of only three $B$ values is as follows. At different $B$ values, MI2 and GT transitions (induced by radiation with $\sigma^+$ or $\sigma^-$ polarization) partially overlap other transitions. At three chosen magnetic field values, partial overlapping...
with other transitions does not occur, which is important for the correct determination of the amplitudes of MI2 transitions marked by 5 and 1− in red circles. According to Fig. 6, the probability of the strongest [2, −2] → [4′, −1′] transition induced by σ+ radiation in a range of 0.2–2 kG is much larger than the probability of the strongest [3, 0] → [1′, −1′] transition induced by σ− radiation, which should be taken into account when using them in magneto-optical processes. Metal atomic vapor is an isotropic medium, but it becomes anisotropic in the applied longitudinal magnetic field B. The different response of atomic systems induced by σ+ and σ− radiation in the magnetic field is called magnetically induced circular dichroism (MICD2). [8, 15]; in this case, it can be called type-2 magnetically induced circular dichroism (MICD2).

We note that we study in this work only nS → nP MI transitions of the first fundamental series of D lines of alkali metals, where n = 3, 4, 5, 6 is the principal quantum number for Na, K, Rb, and Cs, respectively. The total number of these transitions (MI1 and MI2 together) is about 100 transitions, including about 70 MI2 transitions. Calculations show that 70 MI2 transitions will also be observed for the second fundamental series nS → (n + 1)P of D2 lines and for the third fundamental series nS → (n + 2)P of D lines, etc. It is important that the magnetic field B at which the probability of MI2 transitions is maximal decreases for each next series of Dn lines, which will simplify their study and application. It has also been shown that the strongest MI2 transition is induced by σ+ radiation for transitions with ΔF = +2 and has the lowest magnetic sublevel mF for the ground level Fg. Recently fabricated glass nanocells filled with an alkali metal [21, 22], as well as the technical sapphire nanocell used in this work, can be successfully applied for these studies.

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