

Frequency transfer of an optically detected magnetic resonance and observation of the Hanle effect in a nonzero magnetic field

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The method of transferring the frequency of an optically detected magnetic resonance both up and down by an arbitrary value is implemented in a single-beam optical pumping scheme by modulating the linearly polarized beam component. The possibility of observing the Hanle resonance in a magnetic field virtually zeroed upon transition to a rotating coordinate system is demonstrated. A model experiment was carried out, confirming the fundamental feasibility and effectiveness of the method.

Keywords: Optically detectable magnetic resonance, optical resonance frequency transfer, Hanle effect, Bell-Bloom scheme, quantum magnetometer.

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Introduction

In 1924 Wilhelm Hanle published a report [1] on the nontrivial dependence of the polarization of light re-emitted by atoms on the magnitude of the magnetic field, and in 1961 Bell and Bloom for the first time demonstrated [2] the possibility of optical pumping and excitation of magnetic resonance (MR) in atomic medium by means of modulation of the pumping beam. A circularly polarized beam transverse to the magnetic field (MF) induction vector was modulated at a frequency of ω_p . When this frequency coincided with the Larmor frequency $\omega_L = \gamma B_0$ (γ — gyromagnetic ratio, B_0 — induction modulus MF), they registered MP with parameters not worse than in „classical“ scheme, i.e. when exposed to atoms of a radio frequency field with the same frequency. Bell and Bloom proposed the following interpretation of this effect: a beam that turns on only at certain times with a frequency of $\omega \approx \omega_L$ can be considered as stationary not only in the laboratory coordinate system (CS), but also in CS that rotate both clockwise and vs with frequency ω (fig. 1).

We assume that the induction vector MF is directed along the z axis of the laboratory Cartesian CS, the pump beam — along the x axis. In CS $0x'y'z$, which rotates with a frequency of $\omega = \omega_L$ (in the same direction as the atomic magnetic moments), the effective MF acting on the atomic moments is reset to zero: $B'_0 = B_0 - \omega_L/\gamma = 0$, and the pumping of magnetic moments occurs in the direction of beam propagation. It turns out that in a rotating CS, the transverse pumping effect (the Bell and Bloom effect) can be explained in the same way as Hanle himself explained the effect he discovered [1]: in a sufficiently weak magnetic field ($B_0/\gamma = \omega_L < 1/\tau$, where τ — relaxation time) the emitting dipole relaxes before precession has time

to average the parameters of its interaction (absorption, refraction, re-emission) with radiation.

Both effects — and the Hanle effect, and the Bell and Bloom effect allow other interpretations that do not use the classical theory of magnetic moment dynamics in MF. In particular, the most obvious explanation of the Bell and Bloom effect is given by the theory of parametric resonance: by modulating the parameters of a resonant system (for example, the pumping rate), it is possible to cause resonance in this system. The connection of the Bell and Bloom effect with the phenomenon of coherent population captivity is not so obvious, but it is undeniable. Both effects (Bell and Bloom and Hanle) can also be interpreted in terms of interference of atomic states (or, as manifestations of this interference are sometimes called, „of quantum beats“). P.A. Franken [3] pointed out the connection of the Hanle effect with the level crossing effects recorded in nonzero fields in 1961, the interpretation of the Hanle effect within the framework of the theory of interference of atomic states was proposed by E.B. Alexandrov and co-authors in 1965, G. [4], and in 1967, G. Alexandrov proposed the idea of a magnetometer based on the Hanle effect [5]. This idea, being supplemented by the effect of suppressing spin-exchange relaxation under conditions of high concentrations of atoms and high optical pumping rates [6,7], was the basis for the development of the most sensitive zero-field magnetometers SERF (spin-exchange relaxation free) [8].

But the most appropriate to the topic of this article is the interpretation within the framework of the classical dynamics of magnetic moments used by Hanle in [1] and Bell and Bloom in [2] (Fig. 1). The assumption that the equivalence of the magnetic field and the rotation of the coordinate system for an atomic transition — is not just a

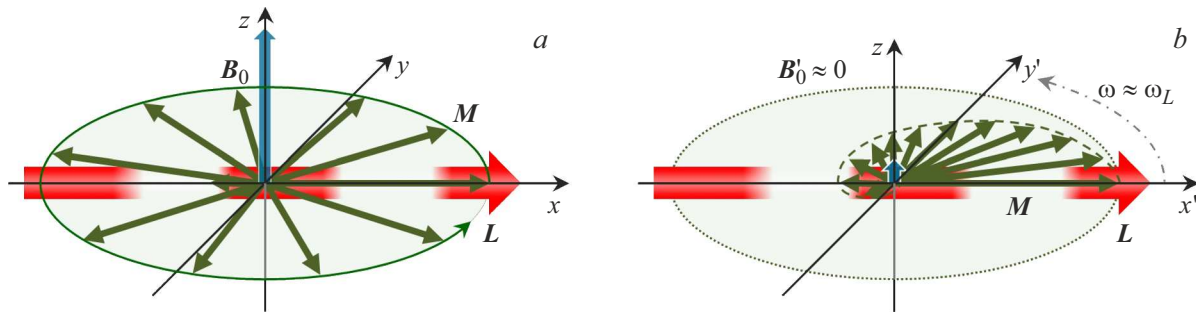


Figure 1. Dynamics of the magnetic moment M of an atomic ensemble in the field B_0 when pumped by a beam L pulse modulated in intensity with frequency ω , close to the Larmor precession frequency ω_L : a — in the laboratory coordinate system $0xyz$; b — in the coordinate system $0x'y'z$ rotating around the axis z with frequency ω . With a sufficiently high modulation duty cycle, the beam can be considered stationary in both CS.

convenient mathematical technique, but a physical reality — eliminates the difference between these effects.

Thanks to the advent of compact single-mode diode lasers, the last two decades have been marked by a sharp increase in interest in quantum sensors with optical pumping [8–14]; the replacement of spectral lamps (also named after Bell and Bloom) with lasers made it possible to modulate any parameters of pumping radiation — intensity, frequency, polarization. Therefore, the Bell and Bloom scheme, which at one time did not become widespread due to limited feasibility, is now widely used in the development of quantum sensors — both magnetometers and gyroscopes. In particular, this scheme makes it possible to exclude radio frequency fields from the sensor circuit, and thereby suppress mutual interference during the operation of sensors in the array; this is critically important for solving the most ambitious tasks of modern quantum magnetometry — creating magnetocardiological and magnetoencephalographic complexes [15–22]. The effect was studied in detail in the works of D. Budker, A. Weis (Weis) and co-authors [23–26], in particular, in [24] the parametric dependences of the Bell and Bloom effect on hyperfine transitions in the ground state Cs were investigated.

In the paper [2] MF was both pumped up and detected at the frequency $\omega_p \approx \omega_L$. In the paper [25], and then [26,27], pumping and detection were performed by a frequency modulated linearly polarized laser beam, and alignment resonances were observed at the pump frequency and double frequency. There, for the first time, the resonance transfer from the zero frequency region was proposed due to the modulation of the pump-detection beam.

In this paper we propose a continuation of these experiments. We propose to shift the frequency of the detected MF by modulating the detecting (test) beam at an arbitrary frequency — both equal to the pumping frequency and different from it. The same logic as described above applies to this case: the detecting beam modulated at the frequency ω_d is stationary in the laboratory CS and in two CS rotating with the frequency ω_d . In this case, the induction of the effective MF is equal to $B'_0 = B_0 \pm \omega_d/\gamma$ (the sign depends

on the direction of rotation of the CS), and the observed frequency of MR is $|\Delta\omega|$ ($\Delta\omega \equiv \omega_p \pm \omega_d$) — it is taken into account here that the forced precession of atomic magnetic moments does not occur at the Larmor frequency ω_L , but at the frequency of the forcing action ω_p .

This conclusion can be considered trivial, since the technique of transferring the frequency of optical signals from the low-frequency region through the use of disk modulators (stroboscopes) was used at the dawn of the development of optics. The advantages of such a transfer are obvious in the problems of measuring ultra-weak and zero MF, in which the frequencies of the recorded signals lie in the domain of dominance of the flicker noise of laser radiation. The modulation of the detecting beam was used to obtain the harmonics of the atomic moment alignment signal in the papers mentioned above [25,26]. The paper [28], is also interesting in this sense, in which the effect of quantum beats was observed by physically rotating the quantum center on the nitrogen-vacancy (NV⁻) center in a diamond.

But, since in the case of MF, the frequency shift can be described not only as a result of banal heterodyning, but also as a consequence of changes in the effective field acting on atoms, we can count not only on practical application, but also on some new interpretations of known physical effects. Thus, the proposed technique is of methodological interest.

In particular, the modulation of the test beam at the frequency $\omega_d = \omega_p \approx \omega_L$ allows you to observe MF at the frequency $\Delta\omega = 0$. Indeed, a probe beam switched on for short periods of time will detect atomic moments in the same precession phase. In another possible interpretation — in a SC rotating with an angular velocity $\omega_d = \omega_L$, the effective MF is zero, and therefore the observed resonance of quantum beats is the Hanle resonance [29], although it can occur in any field values. As befits the Hanle resonance, it is observed when the condition $\Delta\omega < \Gamma$ is met, where $\Gamma = 1/\tau$ — the relaxation rate of magnetic moments, and manifests itself primarily in the rotation of the polarization of the probe radiation at zero frequency.

Problem formulation

The technique of transferring the MR frequency and observing the Hanle resonance in a non-zero field was tested in a single-beam MF sensor built in accordance with the scheme [30] proposed by us earlier. To pump and detect MF in this scheme, a single beam with ellipticity varying in time with the frequency ω_L is used: from the left circular polarization to linear and then — to the right circular. In this case, the circularly polarized component performs optical pumping (according to the Bell and Bloom method), and the linearly polarized — magnetic resonance detection. The measured value is the rotation of the azimuth of linear polarization when interacting with optically oriented atoms. In this paper, we modified this scheme by replacing the sinusoidally time-modulated pulse pumping and decoupling the detection and pumping frequencies.

The signals controlling the ellipticity of the polarization of the pump beam (Fig. 2) can be written as follows:

$$S_1(t) = 2\Theta[\sin(\omega_p t)] - 1, \quad (1)$$

$$S_2(t) = \Theta \left[k_D - \frac{1}{2} - \frac{1}{\pi} \arcsin(\cos(\omega_d t)) \right], \quad (2)$$

where $\Theta(x)$ — Heaviside function: $\Theta(x) = 1$ for $x \leq 0$ and $\Theta(x) = 0$ for $x < 0$. The signal S_1 is responsible for the circular pumping component, the signal S_2 with the fill factor k_D — for the linear one. The degree of ellipticity of polarization at the output of the electro-optical modulator is proportional to the applied voltage. The voltage $U(t) = AS_1(t)(1 - S_2(t))$ is applied to the input of the modulator, where A is a constant coefficient. The coefficient A is chosen such that when $U(t)$ changes in the range from $-A$ to $+A$, the ellipticity changes from -1 to $+1$ (from -45° to $+45^\circ$ in terms of phase delay). If the frequencies ω_p and ω_d satisfy the condition $\omega_d/\omega_p = m/n$, where n and m are — integers, the function $U(t)$ is periodic with the period $m2\pi/\omega_p = n2\pi/\omega_d$. Fig. 2, 3 shows the case when the frequencies differ by 5% ($m = 19$, $n = 20$).

Complex modulation of the pump-detection beam in case of frequency mismatch f_p and f_d lead to the appearance of additional spectral components (Fig. 3, a), including in the vicinity of the resonance frequency (Fig. 3, b): a component appears in the pump spectrum at the frequency $f_p - 2\Delta f$, where $\Delta f \equiv \Delta\omega/(2\pi) = f_p - f_d$ and a significantly weaker component at the frequency $f_p + 2\Delta f$. The resulting MF distortions can be avoided by choosing either multiple frequencies f_p and f_d , or those for which $2\Delta f \ll \Gamma/(2\pi)$.

Experiment

The experimental part of the work was carried out on the installation described in [30,31] and representing a mock-up of a magnetometric sensor with an external pump source placed in the MF stabilizer (Fig. 4).

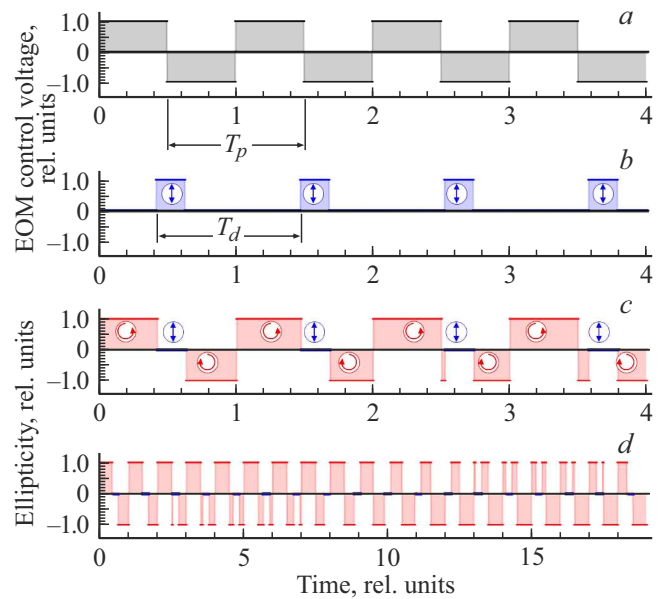


Figure 2. The principle of pump-detection beam formation: *a* — control signal S_1 modulated at the frequency $f_p = \omega_p/(2\pi) = 1/T_p = 1.0$; *b* — control signal S_2 , modulated at the frequency $f_d = \omega_d/(2\pi) = 1/T_d = 0.95$, $k_D = 0.2$; *c, d* — time diagram of the ellipticity of the pump-detection beam at $f_p = 1.0$, $f_d = 0.95$, $k_D = 0.2$; the intervals in which the beam is linearly polarized are highlighted in blue. The arrows show the polarization states of the beam.

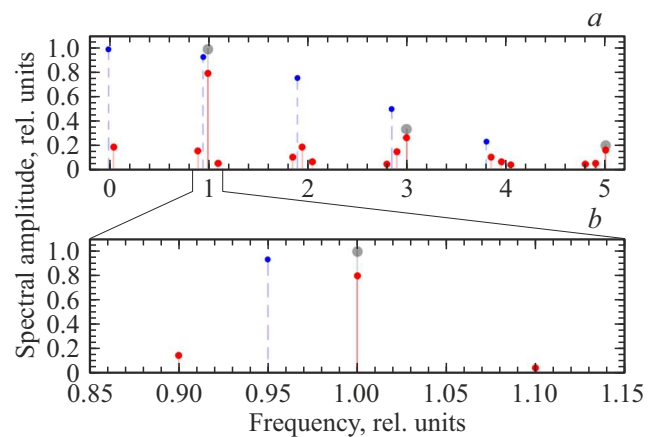


Figure 3. Pump-detection radiation spectrum at $f_p = 1.0$, $f_d = 0.95$: *a* — in the range $(0-5)f_p$; *b* — in the range of $(0.85-1.15)f_p$; the initial spectral amplitude of the pump beam corresponding to Fig. 2, *a* is highlighted in gray; the spectral amplitude of the detection beam corresponding to Fig. 2, *b* is highlighted in blue; the spectral amplitude of the ellipticity of the pump beam in red is highlighted in the presence of a modulated detection beam, corresponding to Fig. 2, *c, d*.

A cubic cell with a size of $8 \times 8 \times 8$ mm contained saturated caesium and nitrogen vapors under pressure of ~ 100 Torr and was placed in the central region of a multilayer magnetic screen, in which the induction of MF in the range of $5-12 \mu$ was maintained using an active

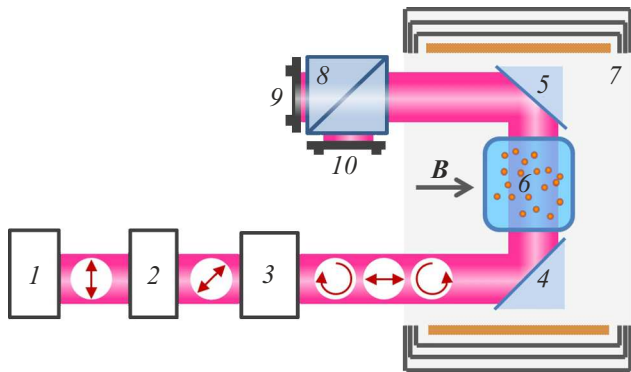


Figure 4. Simplified experimental scheme: 1 — semiconductor laser with external resonator, 2 — optical insulator, 3 — electro-optical polarization modulator, 4, 5 — mirrors, 6 — gas cell with vapors Cs, 7 — magnetic shield with solenoid, 8 — polarizing beam-splitting cube, 9, 10 — photodetectors. The arrows indicate the polarization states of the beam; after the EOM, they correspond to different modulation phases.

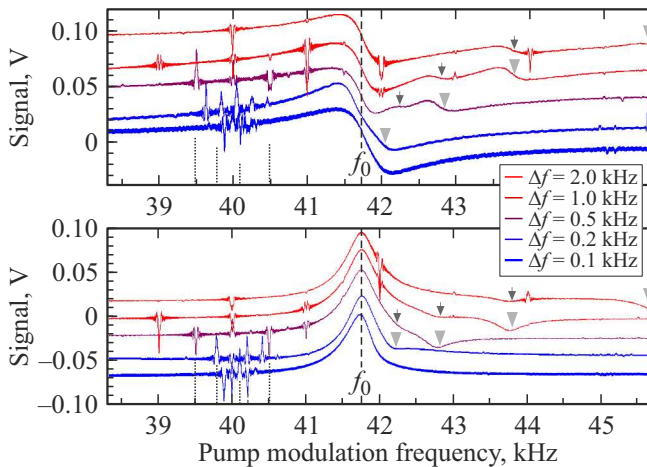


Figure 5. Records of two quadrature components of MR when scanning frequencies f_p and f_d in the vicinity of MR with a constant difference Δf between them. The MR signal was detected at a frequency Δf (0.1–2.0 kHz). The large arrows mark the signal caused by the spectral component at the frequency $f_p - 2\Delta f$, the small arrows — signal caused by the spectral component at the frequency f_d .

stabilization scheme T. The ellipticity of the polarization of laser radiation was modulated using EOM at a constant intensity of this radiation. The measured value was the angle of rotation of the azimuth of polarization of the linearly polarized component of the beam.

Fig. 5 shows the recordings of two quadrature components of the MR when scanning the frequencies f_p and f_d in the vicinity of the MR with a constant difference Δf between them (from record to record Δf varied from 0.1 to 2.0 kHz, synchronous detection of the digitized signal was performed numerically at a frequency of Δf). The amplitude of the down-shifted (heterodyned) signals is

approximately 30% of the original amplitude, which is explained by the redistribution of the signal by harmonics. In addition to the main MR signal, there is a signal due to the spectral component of pumping at the frequency $f_p - 2\Delta f$ (Fig. 3, b), and a very weak signal due to the destructive contribution of the detection beam to pumping at the frequency f_d . The „rattle“ observed in the vicinity of the frequency $40 \text{ kHz} \pm n\Delta f$ (n — integer) is caused by beats with the digitization frequency of the signal (10 kHz).

Similar resonances with similar parameters were observed at frequencies $f_p + f_d$. Thus, we have successfully carried out the transfer of the signal up and down in frequency.

Further, we investigated the possibility of transferring the resonance to a zero frequency, i.e. the possibility of observing the Hanle effect in a non-zero magnetic field (frequency MR $f_L = 20 \text{ kHz}$). The results are shown in Fig. 6, 7. Fig. 6 shows the dependences of the parameters of resonances detected at the zero (Hanle signal), first and second harmonics of the frequency f_d on the fill factor of the test pulse k_D . For optimal values, $k_D = 0.2$ – 0.3 the amplitude of the Hanle signal exceeds the amplitudes of the other signals by 1.5–2 times. The widths of the signals (all of them are given in the scale f_d) do not differ so significantly. The view of the original signal containing all three harmonics is shown in the inset in Fig. 7. Further, with the value $k_D = 0.2$, providing maximum resonance steepness, we varied the difference frequency Δf from zero to 2 kHz ($\Delta f \ll f_L$) and measured two types of response — at zero frequency and at the frequency Δf (Fig. 7).

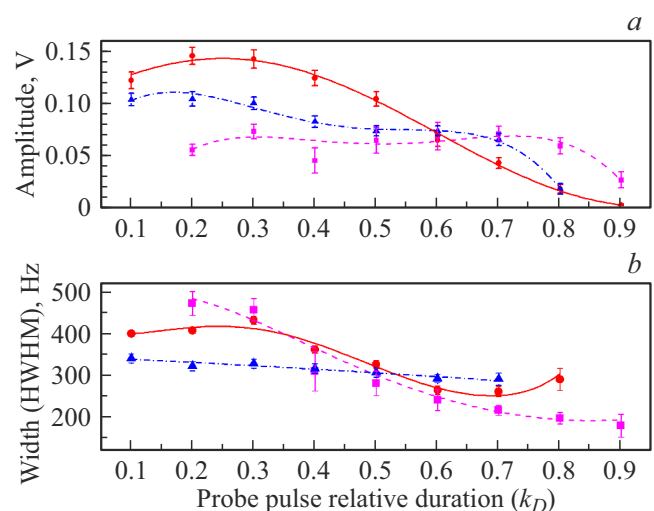


Figure 6. Parameters of the MR signal recorded during frequency scanning f_p and f_d in the vicinity of MR with a constant difference $\Delta f = 0$ between them and the projected: ● — at zero frequency (Hanle resonance), ■ — at frequency f_d , ▲ — at a frequency of $2f_d$; a — amplitude, b — half-width.

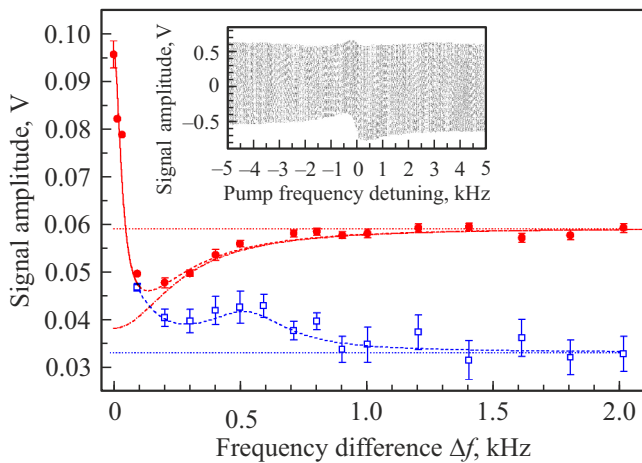


Figure 7. The amplitude of the MR signal recorded during frequency scanning f_p and f_d in the vicinity of MR with a constant difference Δf between them and the projected one: \bullet — at zero frequency, \square — at frequency Δf . Vertical lines — statistical error (3 m.s.d.) obtained by approximation of the resonance by the Lorentz contour. In the inset — recording of the signal while simultaneously scanning the frequencies $f_p = f_d$ ($\Delta f = 0$) in the vicinity of MR; the recording contains the zero, first and second harmonics of the signal.

Discussion

It should be noted some features of the results shown in Fig. 7. First, in addition to the previously discussed signal at the difference frequency Δf (blue dotted line in Fig. 7), as well as the signal recorded at zero frequency at $\Delta f \ll \Gamma/(2\pi)$ (Hanle signal — red solid line in Fig. 7), we registered another signal at zero frequency, characterized at $\Delta f > \Gamma/(2\pi)$ by approximately constant amplitude (the red dashed dot line in Fig. 7). The origin of this signal is still unclear. Possible versions: 1) non-linearity in the registration channel, for example — asymmetric saturation of photodetectors; 2) parasitic pumping of MR, „by the detection beam“, i.e. linearly polarized beam component as a result of ellipticity introduced into it by optical elements; 3) parasitic detection of MR, „by the pump beam“, i.e. circularly polarized component of the beam as a result of the linearity introduced into it. All these effects should lead to the appearance of a signal at zero frequency, characterized by an amplitude almost independent of Δf . However, our measurements show that the ellipticity (or linearity) introduced by the silver mirror does not exceed 1° – 2° , which is clearly insufficient for the occurrence of an effect of this magnitude; the same can be said about the nonlinearity in the registration channel.

Secondly, the width of the MR registered by all the methods listed above remains approximately constant, and in a wide range of parameter changes is 300–400 Hz HWHM, where 300 Hz — own resonance width, and the rest — hardware broadening. But at the same time, the decrease in the amplitude of the MR measured at

zero frequency (Hanle signal — solid line in Fig. 7), when detuning the frequency Δf from zero, it occurs much faster — the peak in the vicinity of $\Delta f = 0$ is characterized by a half-width of only ~ 50 Hz. This is a manifestation of the specifics of our registration method, primarily due to the fill factor k_D (the inverse of the well) of the detecting component of the beam. The usual condition for observing the Hanle signal is formulated as follows [29]: $\omega_L \tau < 1$, where $\tau = 1/\Gamma$ — that is, the atomic ensemble must lose coherence in a time less than that in which it occurs averaging of polarization in all directions in the plane of precession of the atomic moment. With our registration method, the relative duration of the open „window“ detection (determined by the coefficient k_D) imposes an additional restriction: $\omega_L \tau < k_D$, which implies $\Delta f < k_D \Gamma / (2\pi)$ and, therefore, when $k_D = 0.2$ and $\Gamma / (2\pi) = 300$ Hz the boundary frequency is $\Delta f_B \approx .60$ Hz. Once again, we emphasize that the narrowing of the peak in the frequency representation does not lead to a decrease in the MR width — it remains equal to Γ .

Conclusion

We have demonstrated the possibility of transferring the MR frequency in a single-beam pump-detection scheme to an arbitrary frequency domain, and as a special case — the transfer of the MR frequency to the zero frequency by purely optical methods — by modulating the parameters of the detecting radiation. We have shown that such a transfer is possible, including in a single-beam Bell-Bloom scheme that does not use radio frequency fields, and that the result is equivalent to the observation of the Hanle resonance. In addition to purely applied (transfer of the resonance signal to an arbitrary frequency range free from technical noise of laser radiation), this experiment has methodological significance, since it demonstrates the possibility of observing the Hanle effect in an arbitrarily strong magnetic field and once again confirms the connection of the Bell-Bloom scheme and the Hanle effect.

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Conflict of interest

The authors declare that they have no conflicts of interest.

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