Gaskinetic magnetic resonance in the gases N_2 and CO

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Results are presented of an investigation of the resonant change of the thermal conductivity of the gases N_2 and CO in mutually perpendicular (crossed) constant (H) and alternating ($H_\sim$) magnetic fields (gaskinetic magnetic resonance, GMR). The dependences of the GMR on the pressure and on the oscillating field strength, obtained for the gases, agree qualitatively with the theory developed for the case when the alternating field $H_\sim$ constitutes a constant field that rotates about H. The theoretically predicted fine structure, to within 10% of its expected depth, was not observed under the conditions of the experiment performed.

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It was theoretically shown in Ref. 1 that in mutually perpendicular (crossed) constant (H) and alternating ($H_\sim$) magnetic fields there should take place a resonant decrease of the transport coefficients of molecular gases (gaskinetic magnetic resonance—GMR) when the frequency ω of the field $H_\sim$ and the field H are respectively equal to their resonant values ω_r and H_r, which are connected by the relation $\omega_r = \gamma H_r$, where γ is the gyromagnetic ratio of the gas molecules. Satisfaction of this relation means equality of the frequency ω to the precession frequency of the molecules in the field H. Experimental observation of GMR of the coefficient of thermal conductivity in oxygen was reported in Refs. 2 and 3, while in nonparamagnetic gases, with nitrogen as the example, it was reported in Ref. 3. GMR appears against a background of the known effect of the monotonic decrease of the transport coefficients with increasing field H—the Senftleben Beenaker effect (SBE).[4–7]

For diatomic molecules, the SBE is well described by a theory[6] that uses a collision model in which the average collision cross section is

$$\sigma \propto P_2(\widehat{\mathbf{v}, \mathbf{M}}) + P_2(\widehat{\mathbf{v}, \mathbf{M}_1}) + P_2(\widehat{\mathbf{v}', \mathbf{M}'}) + P_2(\widehat{\mathbf{v}', \mathbf{M}_1'})$$

(the P_2 model), where P_2 is the second Legendre polynomial, $\mathbf{v}$, $\mathbf{M}$, and $\mathbf{M}_1$ are the relative velocity and the moments of the two molecules prior to the collision, while $\mathbf{v}'$, $\mathbf{M}'$, and $\mathbf{M}_1'$ are the values after the collision. Using the P_2 model and considering $H_\sim$ to be a rotating field H_1, we can obtain from the general formulas of Ref. 1 the following expression for the shape of the curve of the resonant variation of the thermal conductivity $R(H, H_1, \omega, \nu)$ in the case when the temperature gradient $\nabla T \parallel \mathbf{H}$ (see, e.g., Ref. 8), and $\gamma H_r/\nu \gg 1$:

$$R^{\parallel}(H, H_1, \omega, \nu) = \frac{6}{5} S^{\parallel}_{\text{sat}} \frac{a^2}{a^2+x^2} \left[\frac{a^2}{4a^2+4x^2+1} + \frac{x^2}{a^2+x^2+1} \right], \tag{1}$$

where $a = \gamma H_1/\nu$, $x = (\gamma H - \omega)/\nu$, $S^{\parallel}_{\text{sat}}$ is the change, due to the SBE, of the thermal conductivity at $\mathbf{H} \parallel \nabla T$ and $H \to \infty$ (i.e., when the SBE saturates), and ν is the polarization-relaxation frequency. The value of ν can be determined from the expression $\nu = p\gamma (H/p)^{\parallel}_{1/2}$, where $(H/P)^{\parallel}_{1/2}$ is a constant for a given gas and is equal to the ratio of the field to the pressure at which the SBE reaches a value $(1/2) S^{\parallel}_{\text{sat}}$. Thus, $(H/p)^{\parallel}_{1/2} = 7$ kOe/Torr for nitrogen, and to 7.7 kOe/Torr for carbon monoxide (Ref. 7).

In the case $\nabla T \perp \mathbf{H}$ we have $R^{\perp} = 1/4 R^{\parallel}(H, H_1, \omega, \nu)$. The relative amplitude of the resonance $R_0 = R(H = H_r)/S^{\parallel}_{\text{sat}}$ is equal to

$$R_0^{\parallel} = {}^6/_5 a^2/(4a^2+1), \quad R_0^{\perp} = {}^3/_{10} a^2/(4a^2+1) \tag{2}$$

for parallel and perpendicular geometry, respectively.

According to (1), at values $a > 1/\sqrt{8}$, a fine structure appears at the peaks of the resonance curves. In the general case, the form of the fine structure[1,8] is determined by the chosen model of the collisions, i.e., in final analysis, by the angular dependence of the potential of the interaction of the molecules with one another.

We present below the results of an experimental investigation of the GMR in N_2 and CO. The measurement procedure is similar to that described in Ref. 3. A block diagram of the experimental setup is shown in Fig. 1. Two identical

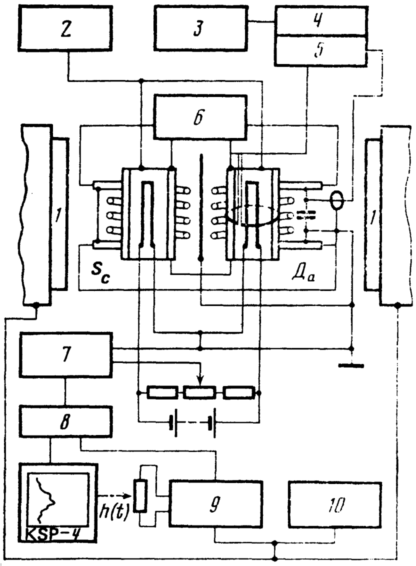

FIG. 1. Block diagram of the experimental setup: 1—electromagnet; 2—system for evacuation and filling; 3—master oscillator Ch6-31; 4—power amplifier; 5—amplitude stabilizer; 6—thermal stabilization system; 7—F116/1 amplifier; 8—synchronous detector; 9—modulator; 10—stabilizer for the field H_0; S_c, S_a—comparison and active sensors.

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heat-conduction sensors, S_a and S_c, similar to those described in Ref. 9, are placed in the gap of the electromagnet that produces the constant field H. Each sensor is a cylindrical glass chamber with inside diameter 18 mm, in the axial cross section of which is placed a thin (3 μm) mica plate (45×16 mm) both sides of which are coated with thermistors of gold in the form of U-shaped strips of width 4 mm, connected in series and bifilarly. The chambers of the sensors communicate with each other and are connected to a system for evacuation and for entry of the investigated gas. The thermistors are connected in adjacent arms of a Wheatstone bridge and are heated by the current that supplies the bridge (the source is a storage battery). The housing of the sensor S_a (active) is placed in a solenoid that serves as the inductance of the output tank circuit of a high frequency oscillator and produces an alternating magnetic field $H_\sim = 2H_1\cos\omega t$, perpendicular to the constant field. The solenoid placed over the sensor S_c (comparison) is short circuited and ensures thermal symmetry and screening of S_c against the stray field of the first solenoid. To decrease the thermal drifts, a system is used for water-cooling the solenoid and thermal stabilization of the sensor walls. The ranges of variations of $H, f=\omega/2\pi$, H_1, and p are respectively 0–2.4 kOe, 183–606 kHz, 60–180 Oe, and 7–30 mTorr.

The measurements were performed by a modulation procedure. To this end, a parallel field of practically rectangular waveform, with an amplitude $H(t)$ that varies linearly with the time t, was superimposed on the constant field $H=H_0$ (Fig. 2). The modulation period was $T\approx 4$ sec, the time required to sweep h from $h=0$ to the maximum value $h=h_m$ was $t_{\text{meas}}\approx 25$ min. The field-induced pulsating bridge-unbalance voltage was amplified and synchronously detected, generating a useful signal ε proportional to the difference between the changes of the thermal conductivities of the gas in the sensors S_a and S_c when the field was varied from $H=H_0$ to $H=H_0+h(t)$. The sensor S_c serves to compensate for the signal due to the SBE, which in our case is an unnecessary background. As a result of this compensation, the signal ε turns out to be proportional in practice only to the resonant change of the thermal conductivity $R(H_0, h)$:

$$\varepsilon \infty R[H_0+h(t)]-R[H_0].$$

In addition, the presence of the sensor S_c decreases the influence of the thermal drifts on the output signal of the measuring bridge. The signal $\varepsilon[H_0, h(t)]$ was recorded with an automatic plotter. In the figures that follow are shown the results of the statistical reduction of plots obtained under identical conditions. The maximum value was $h_m = 340$ Oe, therefore measurements in a wider range of fields H were carried out by successively changing the initial field H_0 and "joining together" the obtained plots.

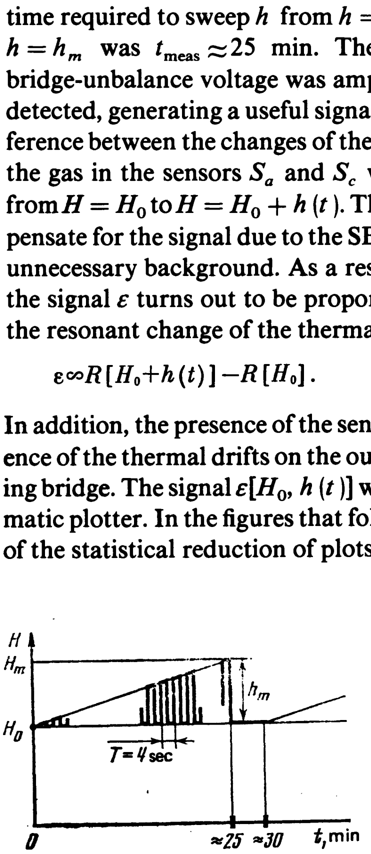

FIG. 2. Time dependence of the modulating field.

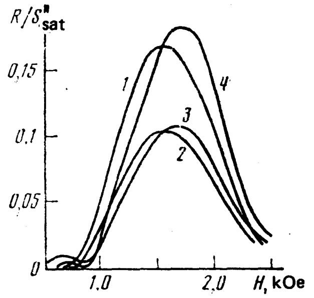

FIG. 3. Experimental curves of GMR in N_2 (1,2) and CO (3,4) at $p=30$ mTorr, $H_1=180$ Oe, $f=338$ kHz, and at two orientations of the temperature gradient ∇T relative to **H**: 1,4 –$\nabla T\parallel\mathbf{H}$; 2,3–$\nabla T\perp\mathbf{H}$.

Figure 3 shows the resonance curves $R(H)/S^{\parallel}_{\text{sat}}$ for N_2 and CO at $\nabla T\parallel\mathbf{H}$ and $\nabla T\perp\mathbf{H}$, $p=30$ mTorr, $H_1=180$ Oe, and $f=3.8$ kHz. It can be seen from the figure that the direction of ∇T does not influence the value of the field H_r: $H_r(N_2)=1.58\pm0.05$ kOe, $H_r(\text{CO})=1.69\pm0.05$ kOe. The values of R_0 for N_2 and CO differ by not more than 10%, which is to be expected in view of the large similarity of the kinetic properties of these gases.[7] For both gases $R^{\parallel}_0/R^{\perp}_0\approx1.8$. This value does not agree with the theoretically expected[2] relation $(R^{\parallel}_0/R^{\perp}_0)_{\text{theor}}=4$. As shown by experiments carried out in a constant field at $H_1=0$ we get $S^{\parallel}_{\text{sat}}/S^{\perp}_{\text{sat}}=0.68\pm0.02$ ($p=30$ mTorr), which differs little (within the framework of the P_2 model) from the theoretical value 2/3. This gives grounds for assuming that the disparity between $(R^{\parallel}_0/R^{\perp}_0)_{\text{theor}}$ and $(R^{\parallel}_0/R^{\perp}_0)_{\text{exp}}$ can only be partially attributed to the inhomogeneity of the field ∇T in the sensor.

Experiments carried out at frequencies 183, 250, 338, and 490 kHz have shown that the resonant values of the field H_r are respectively (0.86; 1.2; 1.6; 2.3) $\pm$ 0.05 kOe for N_2 and (0.95; 1.3; 1.7) $\pm$ 0.05 kOe for the first three frequencies in the case of CO. The gyromagnetic ratios $\gamma^*=\gamma/2\pi=f/H_r$ calculated from those data are equal to $\gamma^*_{N_2}=210\pm10$ Hz/Oe and $\gamma^*_{\text{CO}}=195\pm10$ Hz/Oe, in good agreement with their exact values (Refs. 10 and 11).[1)] The result of the measurements of R_0 when the basic parameters of the experiments are varied are listed in Table I. The relative error in the determination of R_0 is $\sim5\%$. It can be seen from the table that R_0 increases with increasing H_1 and with decreasing p. No dependence of R_0 on ω was observed in the investigated range of values of H_1, p, and ω. The observed dependences of R_0 on these quantities are in qualitative agreement with (2).

In accordance with (1), for a reliable observation of the proposed fine structure of the GMR we chose the following experimental conditions: $\nabla T\parallel\mathbf{H}$, $f=\omega/2\pi=338$ kHz, $H_1=180$ Oe, and $p=30$ mTorr. In this case the saturation of the SBE reaches $\approx95\%$ at $H\approx H_r$ and a value $a=\gamma H_1/\nu\approx0.8$ is ensured, from which, in accordance with the theory, the relative depth of the dip in the fine structure in the GMR is $\delta R/R(H_r)\gtrsim5\cdot10^{-2}$. The measurements were per-

TABLE I. Relative GMR amplitudes in N_2 and CO measured under various experimental conditions.

Gas	∇T	f, kHz	H, Oe	P, mTorr	$10R_0$
N_2	∥	338	180	30	1.7
			100		0.8
	⊥	338	160	15	1.0
			60		0.45
			180	30	0.9
				15	1.15
				7	1.4
CO	∥	338	180	30	1.8
			100		0.8
	⊥	338	180	30	0.95
			100		0.45

formed in the range $\Delta H = h_m = 340$ Oe in the region of the maximum of the resonant dependence. The result of a statistical reduction of eight diagrams obtained for N_2 under identical experimental conditions is shown in Fig. 4. It can be seen that no fine structure of the GMR is observed, and even if it does exist, under our conditions $\delta R / R (H_r) < 5\cdot 10^{-3}$, which is smaller by more than one order of magnitude than the expected value. A similar result was obtained also in the case of CO.

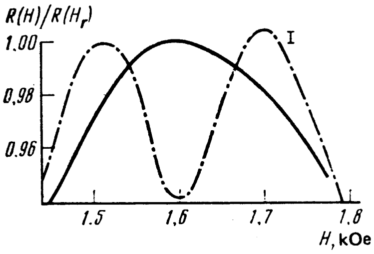

FIG. 4. Shape of GMR curve in the region of the proposed fine structure. Solid line—experimental data for N_2 at $p = 30$ mTorr, $H_1 = 180$ Oe, $f = 338$ kHz. Dash-dot line—theoretical curve. On the upper right is shown the mean-squared error bar.

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[1)] It must be recognized that in the described experiments we used $^{14}N_2$, whereas in Ref. 10 they used $^{15}N_2$.

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