Rotational spectrum of a gaskinetic magnetic resonance in oxygen

V. S. Laz'ko and V. A. Ryabov
I. V. Kurchatov Institute of Atomic Energy, Moscow; All-Union Correspondence Institute of the Food Industry

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The resonant decrease in the thermal conductivity of O_2 in crossed magnetic fields (the gaskinetic magnetic resonance) consists of a set of resonant peaks having a fine structure. These peaks are shown to represent the spectrum of rotational states of O_2.

The resonant decrease in transport coefficients in a static magnetic field H crossed with an alternating field $H_\sim$, the "gaskinetic magnetic resonance," was studied in Refs. 1–4. This resonance has been observed experimentally in several gases, the first of which was oxygen.[2] A theory for the resonance[1] predicts two resonant minima in the thermal conductivity for oxygen: one for molecules for which the projection of the electron spin onto the total angular momentum is $\sigma = \pm 1$ and another for $\sigma = 0$. The results of Ref. 2 are at odds with this theory. Subsequent studies by V. D. Borman, L. L. Gorelik, V. S. Laz'ko, B. I. Nikolaev, V. V. Sinitsyn, and V. I. Troyan have

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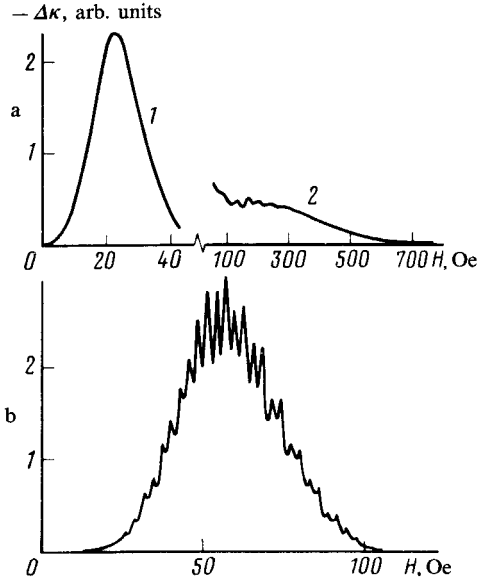

FIG. 1. Resonant dependence of the decrease in the thermal conductivity of O_2. a—$f = 3.176$ MHz, $p = 30$ mtorr, $H = 0.5$ Oe (curve 2 was recorded at an instrumental sensitivity roughly 20 times greater than that for curve 1); b—$f = 7.889$ MHz, $p = 15$ mtorr, $H_\perp = 0.4$ Oe.

revealed a resonance in the thermal conductivity of O_2 which corresponds quite accurately to a gaskinetic magnetic resonance for $\sigma = \pm 1$. The corresponding resonance for $\sigma = 0$ could not be found. In the present letter we report a study of the resonance in O_2 and the discovery of some interesting new aspects of this resonance.

The experimental apparatus and procedure are described in detail in Ref. 4. The procedure[4] makes it possible to discriminate the useful signal, which is proportional to the resonant change in the thermal conductivity, $\Delta\kappa$. We use an alternating field $H_\sim = 2H_\perp \sin 2\pi ft$, where $2H_\perp$ is the amplitude of the field, f is its frequency, and t is the time.

Figure 1a shows a plot of $-\Delta\kappa(H)$ at the frequency $f = 3.176$ MHz. The resonant field value, $H_r = 23.0 \pm 0.5$ Oe, corresponds to a gyromagnetic ratio $\gamma^* = \gamma/2\pi = f/H_r = (1.38 \pm 0.03) \times 10^5$ Hz/Oe, in agreement with the results found by the investigators listed above and with the theoretical value of 1.37×10^5 Hz/Oe at $T = 350$ K for O_2 molecules with $\sigma = \pm 1$. The height of the resonance corresponds to a relative change $\Delta\kappa/\kappa \sim 10^{-3}$. It can be seen from Fig. 1 that the gaskinetic magnetic resonance for $\sigma = 0$ is not observed in the predicted field range, $H = (10\text{–}20)H_r$. When the sensitivity of the apparatus in the region $H = 100$–800 Oe is increased by a factor of about 20, we do see some deviation from a monotonic behavior (curve 2), but a change in frequency does not result in the necessary proportional shift of these nonmonotonic features along the H axis. These features may be due to a recently discovered oscillation effect.[5,6]

Figure 1b shows $-\Delta\kappa(H)$ at the frequency $f = 7.889$ MHz. Here we see a set of peaks whose separation (ΔH) along the H scale is constant, within the errors. We find the ratio $f/\Delta H = (2.79 \pm 0.03) \times 10^6$ Hz/Oe, regardless of the frequency.

The rotation of the homonuclear molecule O_2^{16} with a zero nuclear spin is described[7] by odd values of the rotational quantum number K. The gyromagnetic ratio of

O_2 molecules with $\sigma = \pm 1$ has, because of Zeeman splitting, two values for each specific value of K (Hund's case b) (see, for example, Ref. 8): $\gamma = 2\mu_B/\hbar K$ for $\sigma = -1$ and $\gamma = -2\mu_B/\hbar(K+1)$ for $\sigma = +1$, where μ_B is the Bohr magneton, and $\hbar$ is Planck's constant. The sign of γ (or that of σ) is not distinguished in our experiments, because of the symmetry of the experimental conditions: We use an oscillating field instead of a rotating field, and furthermore the temperature gradient $\vec{\nabla} T$ at the detectors was in opposite directions on the two sides of the thermistors.[4] Consequently, to each value of K there should correspond a pair (doublet) of resonant peaks separated by $\Delta H = hf/2\mu_B$, where $h = 2\pi\hbar$. For all possible odd values of K, these doublets form a sequence (spectrum) of peaks with the same interval ΔH, in complete agreement with Fig. 1b (the peak doublets are particularly noticeable on the slopes of the resonant curve). The ratio $f/\Delta H = 2\mu_B/h = 2.799 \times 10^6$ Hz/Oe agrees with the experimental value.

In the theory for the gaskinetic magnetic resonance,[1] the rotation of the molecules is treated classically, since the average value $K \sim 10$ is quite high at room temperature. The expression for the resonant decrease in the thermal conductivity of oxygen, which was derived in Ref. 1, gives a satisfactory description of the experimental results if we replace the integration over the angular momenta M in it by a summation. This expression (for the case in which the precession frequency $\gamma^* H_\perp$ is considerably larger than the typical relaxation frequency ν),

$$-\Delta\kappa \propto H_\perp^2 \sum_n \Phi_n \frac{4(H - \omega/\gamma_n)^2 + H_\perp^2}{[(H - \omega/\gamma_n)^2 + H_\perp^2]^2}, \tag{1}$$

takes the form of a sum of resonant terms with a weight function $\Phi_n = M_n^5 \exp(-M_n^2/2IkT)$, where k is the Boltzmann constant, I is the moment of inertia, $M_n \approx n\hbar$, $\omega = 2\pi f$, $\gamma_n = 2\mu_B/\hbar n$, and $n = K$ or $K+1$. Here we have omitted the terms with $\gamma = -2\mu_B/\hbar K(K+1)$ for $\sigma = 0$, and we have allowed for the fact that the sign of γ is not determined experimentally.

Figure 2 shows some normalized experimental heights of the resonant peaks, R_n, versus the number $n = H_n/\Delta H$ (H_n is the resonant field for peak n) for various values of $H_\perp$, along with the correspondingly normalized function Φ_n (the solid curve). We see that the behavior is the same in the two cases, confirming the argument above. The spectrum of the gaskinetic magnetic resonance in O_2 can thus be thought of as a

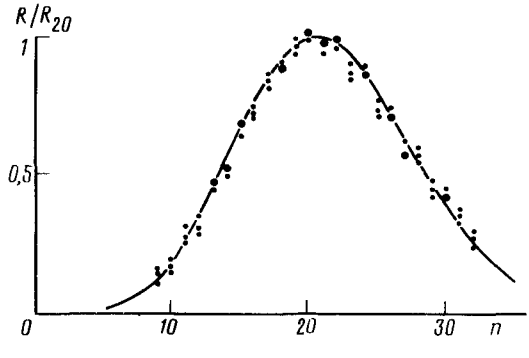

FIG. 2. The normalized experimental heights of the resonant peaks, R_n, versus the peak index n for the three values $H_\perp = 0.2$, 0.6, and 1.0 Oe. The large circles represent cases in which two values of R_n for different $H_\perp$ coincide.

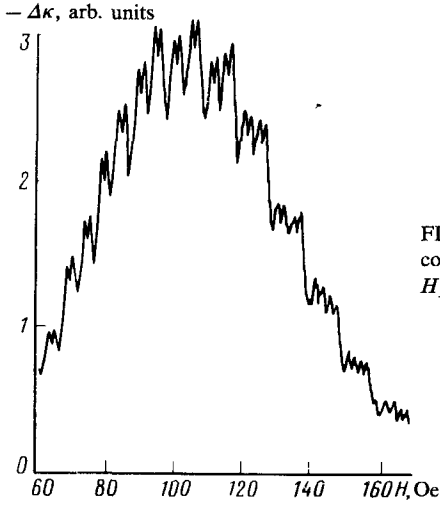

FIG. 3. Resonant dependence of the decrease in the thermal conductivity of O_2 ($f = 15.096$ MHz, $p = 60$ mtorr, $H_\perp = 1.1$ Oe).

superposition of the gaskinetic magnetic resonances involving "nonparamagnetic" components ($\gamma_n = \text{const}$) of the gas. At the average temperature $T = 350$ K in the experiments, the maximum height of the resonant spectrum corresponds to $n = 20$. Since the functional dependence $R_n(n)$ is directly related to the angular-momentum distribution of the gas molecules, sufficiently precise measurements of this dependence might furnish direct information on the deviation of the gas distribution function from equilibrium in angular-momentum space caused by a deviation from equilibrium in velocity space (in the presence of a gradient $\vec{\nabla} T$, for example).

Figure 3 shows the functional dependence $-\Delta\kappa(H)$ found at the frequency $f = 15.096$ MHz. We see that at the crest of each peak it is possible to make out a slight dip. This dip corresponds to the fine structure predicted in the gaskinetic magnetic resonance by the theory: Each resonant term in (1) describes a peak whose crest is split in two at $H = H_n \pm H_\perp/\sqrt{2}$. This type of fine structure of a resonance corresponds to the molecular-scattering model discussed in Ref. 1. The relative depth of the dip, $\delta R/R$, observed experimentally, however, is smaller than the theoretical prediction by a factor of about five. No fine structure could be found in the gaskinetic magnetic resonance in N_2 or CO in Ref. 4. It should be noted in this connection that the ratio $\gamma^* H_\perp/\nu$ in Ref. 4 ($\delta R/R$ increases as this ratio increases) was no more than 0.85, while Fig. 3 yields a value ~ 3. The actual relaxation frequency ν is apparently higher than the theoretical prediction (Ref. 4), approximately equal to the collision rate.

The behavior of the amplitudes R_n as functions of $H_\perp$ and the pressure p agrees qualitatively with (1).

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[1]V. D. Borman, L. A. Maksimov, Yu. V. Mikhaĭlova, and B. I. Nikolaev, Zh. Eksp. Teor. Fiz. **53**, 2143 (1967) [Sov. Phys. JETP **26**, 1210 (1968)].
[2]V. D. Borman, L. L. Gorelik, B. I. Nikolaev, and V. V. Sinitsyn, Pis'ma Zh. Eksp. Teor. Fiz. **6**, 945 (1967)

[JETP Lett. **6**, 362 (1967)].
[3]V. D. Borman, L. L. Gorelik, V. S. Laz'ko, B. I. Nikolaev, and A. V. Khmelev, Preprint IAÉ-2623, Kurchatov Institute of Atomic Energy, Moscow, 1976.
[4]L. L. Gorelik and V. S. Laz'ko, Zh. Eksp. Teor. Fiz. **84**, 931 (1983) [Sov. Phys. JETP **57**, 541 (1983)].
[5]L. L. Gorelik, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 403 (1981) [JETP Lett. **33**, 387 (1981)].
[6]N. S. Averkiev and M. I. D'yakonov, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 196 (1982) [JETP Lett. **35**, 241 (1982)].
[7]G. Herzberg, Molecular Spectra and Molecular Structure, Vol. 1, Spectra of Diatomic Molecules, Van Nostrand, New York, 1939 (Russ. transl. IL, Moscow, 1949).
[8]L. D. Landau and E. M. Lifshitz, Teoreticheskaya fizika, Vol. III, Moscow, 1963 (Quantum Mechanics: Non-Relativistic Theory, Addison-Wesley, Reading, Mass., 1965).

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