

Resonance structure in the magnetic-field dependence of the kinetic coefficients of a paramagnetic gas

N. S. Averkiev and M. I. D'yakonov

A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad

(Submitted 25 December 1981)

Pis'ma Zh. Eksp. Teor. Fiz. **35**, No. 5, 196–198 (5 March 1982)

The dependence of the kinetic coefficients of a paramagnetic gas on the magnetic field under conditions corresponding to saturation of the Senftleben effect should reveal many sharp peaks, caused by intersections of Zeeman levels.

PACS numbers: 51.60. + a

This paper was stimulated by Gorelik's report¹ of the observation of an unexpected and extremely unusual effect: oscillations in the thermal conductivity of gasses (H_2 and O_2) in a magnetic field of 1–2 kOe at room temperature. The relative oscillation amplitude was on the order of 10^{-5} in the O_2 , and the oscillation period was a few tens of oersteds.

In this letter we will examine a new effect which may possibly explain the experimental results of Ref. 1: a resonant change caused in the kinetic coefficients of a paramagnetic gas by level intersections in a magnetic field. The thermal conductivity of a gas decreases in a magnetic field (the Senftleben effect) because the angular momenta of the molecules precess around the field direction during the time intervals between collisions and because the scattering cross section depends on the relative orientation of the angular momentum and the velocity of a molecule.² As the magnetic field is strengthened, the splitting of the levels becomes nonequidistant; in classical mechanics, this would mean that the gyromagnetic ratio begins to depend on the orientation of the angular momentum with respect to the magnetic field and on the strength of the field.³ At even stronger fields (on the order of 1 kOe for O_2), there are many intersections of the Zeeman fine-structure levels. Each intersection corresponds to a vanishing of the precession frequency of molecules whose angular momenta make the appropriate angle with the field. For these molecules, the situation becomes the same as in a zero field, and for this reason there should be a resonant increase in the thermal conductivity at magnetic fields corresponding to level intersections.

Figure 1 shows the Zeeman level scheme of oxygen for the rotational quantum number $K = 3$ (actually, values $K \sim 10$ are important at room temperature). In the absence of a field there is a triplet for each K , with $\lambda = 6 \times 10^4$ MHz, while Δ_K is much smaller: $\Delta_K \approx (0.6K - 2.7) \times 10^3$ MHz (for $K > 7$). In a magnetic field satisfying the condition $\mu_0 H \ll \lambda$ (μ_0 is the Bohr magneton), the splitting of the levels with the total angular momenta $J = K \pm 1$ is linear in the field with g factors $g_{K+1}^{(K)} = 2/(K+1)$ and $g_{K-1}^{(K)} = -2/K$. The splitting of these levels remains essentially linear

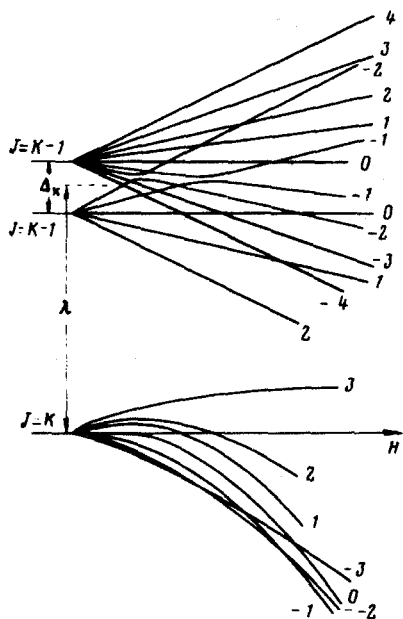


FIG. 1. Zeeman splitting of the fine-structure levels in the O_2 molecule.

even at $\mu_0 H \sim \Delta_K$, so that many intersections of Zeeman sublevels occur. Exceptional cases are the sublevels with identical angular-momentum projections on the field direction, m ; these sublevels undergo "anti-intersections" (Fig. 1), in which the levels approach each other to a minimum separation $\delta_{Km} = (\Delta_K^2/4\lambda) [K(K+1)/m^2 - 1]$. The linear splitting of the $J=K$ level is small [$g_K^{(K)} = 2/K(K+1)$], so that even in comparatively weak fields the perturbation of the Zeeman structure of this level by the interaction with the $J=K \pm 1$ levels becomes important. At $\mu_0 H \ll \lambda$, the splitting of the $J=K$ level is described for $K \gg 1$ by³

$$E_{Km} = \frac{m}{K^2} (2\mu_0 H) - \left(1 - \frac{m^2}{K^2}\right) \frac{(2\mu_0 H)^2}{\lambda}. \quad (1)$$

At $J=K$, there are level intersections at the magnetic field values $2\mu_0 H = -\lambda/(m_1 + m_2)$. The sublevels with $J=K \pm 1$ intersect at $2\mu_0 H = -K\Delta_K/(m_1 + m_2)$, except in the case $m_1 = m_2$. Both of these formulas hold for $|m_1 + m_2| \gg 1$. For oxygen, with $m_1 \sim m_2 \sim K \sim 10$, both types of intersections occur in fields on the order of 1 kOe.

Each intersection should lead to a resonant change in the kinetic coefficients. The Senftleben effect might be said to result from an intersection of all the Zeeman sublevels in a zero field. The total number of these sublevels is on the order of \bar{K}^2 , where \bar{K} is the average thermal value of K . It is thus obvious that the effect with which we are concerned here is of order $1/\bar{K}^2 \sim 10^{-2}$ with respect to the Senftleben effect.

As an example we will calculate the thermal conductivity of a paramagnetic gas from a simple model in which there is a relaxation time τ which depends on the rela-

tive orientation of the rotational angular momentum, \mathbf{K} , and the velocity of the molecule, \mathbf{V} :

$$1/\tau = (1/\tau_0)(1 + \alpha\Gamma), \quad \Gamma = (\mathbf{K}\mathbf{u})^2/K^2 - 1/3, \quad (2)$$

where \mathbf{u} is a unit vector along \mathbf{V} , and α is a parameter describing the extent of the deviation from a spherical geometry. We assume that this parameter is small.

We write the density matrix for the gas as

$$\rho_{ab}(K, \mathbf{V}) = \rho_K^{(0)} \delta_{ab} + (\tau_0 \mathbf{V} \nabla \cdot T/T) g_{ab}(K, \mathbf{V}), \quad (3)$$

where the first term is the equilibrium density matrix, and a and b represent the quantum numbers ($J_1 m_1$) and ($J_2 m_2$), respectively, for a given K . The linearized kinetic equation for g_{ab} is

$$(E(K, \mathbf{V})/T - 7/2) \rho_K^{(0)} \delta_{ab} + (1 + i\Omega_{ab}^{(K)}/\tau_0) g_{ab} + (\alpha/2) (\hat{\Gamma} \hat{g} + \hat{g} \hat{\Gamma})_{ab} = 0. \quad (4)$$

Here $E(K, \mathbf{V})$ is the energy of a molecule with velocity \mathbf{V} and rotational quantum number K , $\Omega_{ab}^{(K)}$ is the difference between the energies of the a and b states for a given K , and $\hat{\Gamma}$ is the operator corresponding to (2). In Eqs. (3) and (4) we have ignored the energies of the spin-orbit and Zeeman splittings everywhere except in $\Omega_{ab}^{(K)}$. The thermal conductivity tensor $\hat{\kappa}$ can be expressed in terms of g :

$$\kappa_{ij} = -(\tau_0/T) \int d^3V V_i V_j \sum_{Ka} g_{aa}(K, \mathbf{V}) E(K, \mathbf{V}). \quad (5)$$

For simplicity, we will restrict the calculations to $\kappa = \text{Sp } \hat{\kappa}/3$ ($\text{Sp} = \text{spur} = \text{trace}$), which determines the total heat flux from a point source.

The effect of interest here, like the Senftleben effect, arises in second order in the parameter α . Solving Eq. (4) by successive approximations, and using (5), we can find the increment $\Delta\kappa$ in the thermal conductivity, which is proportional to α^2 :

$$\frac{\Delta\kappa}{\kappa} = \frac{2\alpha^2}{21\overline{K^2}} \sum_K q^2 (2q^2 + 3) e^{-q^2} \sum_{ab} \frac{\langle |\Gamma_{ab}|^2 \rangle}{1 + (\Omega_{ab}^{(K)}/\tau_0)^2}, \quad (6)$$

where $q = K/\overline{K}$, and the angle brackets denote an average over the orientation of \mathbf{u} . The matrix elements Γ_{ab} are nonzero at $m_1 = m_2$, $m_2 \pm 1$, $m_2 \pm 2$. In the region $\mu_0 H \ll \Delta_K$, λ/\overline{K} , the sum over a and b in (6) becomes

$$\frac{8}{(15)^2} (2K+1) \sum_{J=K-1}^{K+1} \left(\frac{1}{2} + \frac{1}{1 + (\Omega_J^{(K)}/\tau_0)^2} + \frac{1}{1 + (2\Omega_J^{(K)}/\tau_0)^2} \right), \quad (7)$$

where $\Omega_J^{(K)} = g_J^{(K)} \mu_0 H/\hbar$ is the precession frequency in the (K, J) state. Equation (6) describes the Senftleben effect.

Upon level intersections, one of the frequencies $\Omega_{ab}^{(K)}$ vanishes, giving rise to a resonance peak in the thermal conductivity; this peak can be described by the corresponding term in Eq. (6). Anti-intersections of the $(K+1, m)$ and $(K-1, m)$ levels lead to the same effect if $\delta_{Km} \tau_0$ is not too large. The matrix elements Γ_{ab} , which determine the heights of the peaks, can be calculated for $J=K$ through the use of the unperturbed wave functions (J, M) . For $J=K \pm 1$ we must take into account the

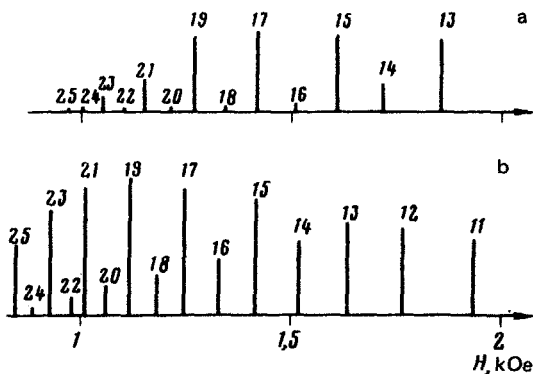


FIG. 2. Positions and relative heights of the resonance peaks in the thermal conductivity for $K = 13$. The peaks are labeled with the values of $|m_1 + m_2|$ for the levels involved in the intersections. a- $J=K \pm 1$; b- $J=K$.

mixing of states at anti-intersections. Figure 2 shows the positions and relative heights of the resonance peaks calculated for both types of intersections for $K = 13$.

The quantity $1/\tau_0$ is on the order of 2 MHz/Torr. Taking into account the slopes at which the various intersections occur, we can find the widths of the peaks along the magnetic-field scale. For $J=K$ and $m_1 - m_2 = \Delta m$ we find $\Delta H \sim K^2 / \Delta m$ Oe/Torr; for the levels with $J=K \pm 1$, the width is far smaller: $\Delta H \sim K / |m_1 + m_2|$ Oe/Torr.

In summary, level intersections in a magnetic field should give rise to a rich spectrum of resonance peaks in the thermal conductivity (and in other kinetic coefficients). The range of magnetic fields, the characteristic distances between adjacent peaks, and the amplitude of the effect predicted by these calculations all agree with the data of Ref. 1 for O_2 . In Ref. 1, however, an oscillation effect was also observed for hydrogen, in the same field range, but with an amplitude two orders of magnitude smaller. Since no level intersections occur for the H_2 molecule in fields ~ 1 kOe, this experimental result cannot be explained by the theory outlined here. The effect might be a consequence of a slight oxygen impurity ($\sim 1\%$) in the hydrogen. This theory also fails to explain the observed dependence of the effect, particularly of the oscillation period, on the pressure. It should be noted, however, that the resonance peaks predicted in the present letter at low pressures should be very narrow, and a highly uniform field would be required in order to study them experimentally.

This analysis shows that subtle features of the energy spectrum of a molecule may be manifested in kinetic phenomena.

1. L. L. Gorelik, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 403 (1981) [JETP Lett. **33**, 387 (1981)].
2. Yu. Kagan and L. A. Maksimov, Zh. Eksp. Teor. Fiz. **41**, 842 (1961) [Sov. Phys. JETP **14**, 604 (1962)]; **51**, 1893 (1966) [**24**, 1272 (1967)].
3. L. A. Maksimov, Zh. Eksp. Teor. Fiz. **61**, 604 (1971) [Sov. Phys. JETP **34**, 322 (1972)].

4. C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy*, McGraw-Hill, New York, 1955
(Russ. transl. IIL, Moscow, 1959).

Translated by Dave Parsons

Edited by S. J. Amoretty