Experimental observation of oscillatory structure in a gaskinetic magnetic resonance

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Oscillatory changes in the intensity of the gaskinetic magnetic resonance have been observed experimentally in a nonparamagnetic gas, specifically, N_2^{14}, upon a change in the static magnetic field. The relative oscillatory change in the resonance in the thermal conductivity is $\sim 10^{-6}$–10^{-5}, and the period of the oscillations is $\sim$10–15 Oe. The observed effect can be linked with a quantization of the angular momenta and a nucleus-rotation interaction.

A resonant decrease ΔK_r occurs in the thermal conductivity of gases in mutually perpendicular static and rf magnetic fields H_- and $H_\sim$. This decrease reaches a maximum magnitude when the precession frequency of the magnetic moments of the molecules in the field $H_- = H_r$ is equal to the frequency (f) of the field $H_\sim$ (a gaskinetic magnetic resonance, GMR).[1,2] Experiments of this type with N_2^{14} have revealed that at given values of $H_\sim$ and f a monotonic change in H_- gives rise to small oscillatory changes in the thermal conductivity, $\Delta K_{0s,r}$, against the background of the change $\Delta K_r(H_-)$ as $H_- = H_r$ is approached. In this letter we report some experimental results (which were first reported in Ref. 3) which reveal this previously unpredicted oscillatory structure in the GMR.

The experimental apparatus and procedure are described in detail in Refs. 4 and 5; certain features of the procedure were reflected in Ref. 3. In the course of the experiments, we record diagrams of the deviation from balance of a measurement bridge as a function of the current through an electromagnet, $U(J)$. The signal $U(J)$ is the sum of the monotonic signal due to the incompletely cancelled background and the signal $U_{0s,r}(J)$ caused by the oscillatory structure of the GMR. The background is caused by a Sönftleben-Binaker effect and the GMR effect.[3] The signal $U_{0s,r}(J)$ is found by subtracting the ordinates of the $U(J)$ curve from the ordinates of the envelope curve, which has no minima. Within $\sim$10%, the value $J = 1$ A corresponds to $H_- = 430$ Oe. All the experiments were carried out at room temperature, most at a pressure $p \sim 30$ mtorr, at $J \sim 1.5$–2 A, at $H_\sim \sim 120$ Oe, and at $f \sim 150$–185 kHz.

Figure 1 shows the relative oscillatory change $U_{0s,r}(J)$ at a pressure $p \sim 30$ mtorr and at $f = 156$ kHz in a range of J corresponding to a variation of H_-/H_r over the

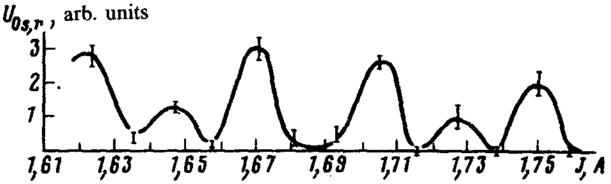

FIG. 1.

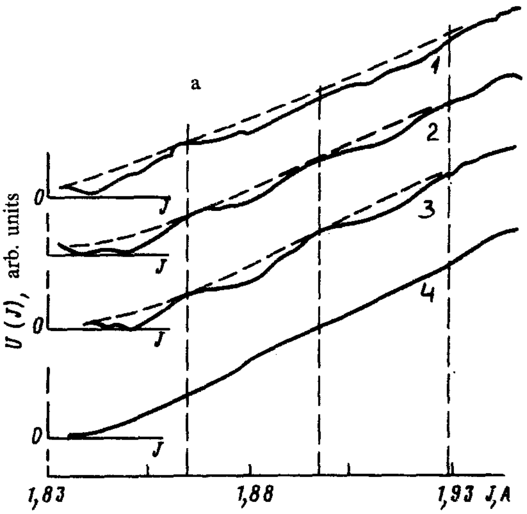

FIG. 2.

interval 0.94–1.03. We see that the oscillation period ΔH_{0s} is ~10–15 Oe. Rough estimates put the peak value of the ratio $\Delta K_{0s,r}/K$ at $\sim 10^{-6}$–10^{-5} (i.e., on the order of a fraction of 1% of $\Delta K_r/K$). Interestingly, oscillations were also observed as the pressure was increased to ~0.6 torr, at which we have $H/P \sim 1$ kOe/torr, i.e., under conditions extremely different from the limiting condition for the appearance of the GMR considered in the theory of Ref. 2. Curves 1–3 in Fig. 2a show three $U_r(J)$ diagrams found in one of these experiments, against the background of the residual, incompletely cancelled Sönftleben-Binaker effect for $J \sim 1.6$–1.95 A and $f = 183$ kHz. Comparison of these curves with curve 4 in diagram 2a ($H_\sim = 0$) shows that under these conditions the GMR consists of distinct, completely resolved peaks. The results of a statistical analysis of several diagrams of this type are shown in Fig. 2b as curve of $U_{0s,r}(J)$; here ΔH_{0s} and $\Delta K_{0s,r}/K$ are ~10–15 Oe and $\sim 10^{-6}$, respectively.

The oscillatory structure of the GMR differs qualitatively, in the way in which it appears, from the oscillatory structure which arises in a static magnetic field (at $H_\sim = 0$): the "oscillatory" effect, described for the particular case[1)] of H_2 in Ref. 3. The period and intensity of the oscillations of the GMR in the case of N_2 are about an order of magnitude greater than for the oscillatory effect in the same gas. The observed oscillatory structure cannot be explained by the existing theory for the GMR.[2] It is reasonable to suggest that the oscillatory structure in the GMR is related to an effect on the GMR of internal molecular fields which are ignored by this theory. The interaction of the internal field with the angular momentum (M) of the molecule causes the precession frequency (ω) of the angular momentum to change by an amount $\Delta\omega \sim E/M$, where E is the energy transfer to it. In the N_2^{14} molecule the angular momentum interacts with the nuclear quadrupole moments through the gradient of the electric field produced by the electron cloud.[7] In this case, oscillations can arise in connection with the quantization of E and M. This interpretation is supported, in particular, by data obtained on resonant spectra by a molecular-beam method.[7–9] Since an initial premise of the theory in this case, as in the case of the GMR, is a resonant nature of the precession of the angular momenta in crossed fields,[8] it seems completely legiti-

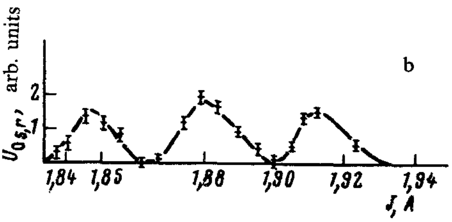

FIG. 3.

mate to pursue the analogy in the interpretation of the oscillations obtained by these methods (despite the fact that they correspond to different physical manifestations of precession).

Studies of the oscillatory structure of the GMR can create new opportunities for studying the structure of molecules. For example, in contrast with the method of the oscillatory structure of the GMR, the sensitivity of the molecular-beam method is inadequate for detecting a resonant spectrum in N_2^{14} (Ref. 7). Since the GMR can be observed at relatively low values of H_{-}/P and $H_{\sim}/P$, as it turns out, there is the possibility of substantially simplifying the GMR experimental procedure. It appears that, after certain refinements are made in the method of the GMR oscillatory structure, and after a corresponding theory has been derived, this method may prove useful for studies of many types.

A well-defined GMR oscillatory structure has recently been observed for O_2 with this apparatus by V. S. Laz'ko. This structure is associated with a quantization of the angular momentum and its interaction with a net electron spin of the electron cloud (which determines the paramagnetism of this gas).[10] However, the applicability of the results found for O_2 in the development of new research methods is basically limited to paramagnetic gases. A further limitation is that in this case the sensitivity of the GMR method is much poorer than that of the electron paramagnetic resonance method.[2)]

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[1)] An oscillatory effect has also been observed in N_2, CO, and D_2. It has been shown that, in contrast with the suggestion in Ref. 6, this effect is not due to oxygen impurities.

[2)] Since all the gaskinetic effects in fields are of "rotational" origin, the term "oscillation" structure (or spectrum), which has become the conventional term, and which was proposed for describing such effects by I. K. Kikoin, should be used in place of the term "rotational" GMR spectrum, which was used in Ref. 10, in order to be more specific in conveying the nature of the structure and for uniformity in terminology. In particular, this usage would reflect the distinction between the oscillatory structure of the GMR and other nonmonotonic structural features of GMR (e.g., the central minimum of the GMR and GMR harmonics).

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