Investigation of the low-frequency antiferromagnetic resonance branch in NiF_2

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The low-frequency antiferromagnetic resonance (AFMR) branch is investigated experimentally in tetragonal weakly magnetic NiF_2 in the 0.6–3.0 mm wavelength range in magnetic fields of up to 130 kOe at a temperature of 4.2°K. The gap in the low-frequency branch of the spin wave spectrum is determined in a zero magnetic field and is found to equal 112 ± 1 GHz. It is found that at frequencies less than ~ 170 GHz the dependence of the resonant magnetic field strength H_r on the direction of the magnetic field **H** lying in the (001) plane can be described within the limits of experimental accuracy by the relation $H_r(\varphi)/H_r(O) = 1/\cos\varphi$ $(O \leq \varphi \leq 45°)$, where φ is the angle between **H** and the [100] direction. At larger frequencies this relation is not satisfied. The dependence of the frequency ν of the AFMR low-frequency branch on the magnetic field strength is investigated for two directions of the magnetic field, **H** along [100] and **H** along [110]. As a result of a least-squares analysis of the experimental data it is found that the function $\nu = \nu(H)$ can be expressed as $\nu^2 = AH^2 + BH + C$, where the coefficient values are given in (3) for **H** along [100] and in (4) for **H** along [110].

INTRODUCTION

At the present time an intensive investigation of the weak ferromagnetism of antiferromagnets is being carried out by a variety of methods. The theory thereof has been suggested by Dzyaloshinskiĭ[1] on the basis of the Landau thermodynamic theory.

We examine the effect of weak ferromagnetism on the high-frequency properties of antiferromagnets in which the magnetizations of the sublattices and also the spontaneous ferromagnetic moment in the absence of a magnetic field lie in a plane perpendicular to the principal crystal axis (i.e., in the basal plane). These conditions are fulfilled, for example, by antiferromagnets with rhombohedral (D_{3d}^6) crystallographic structure ($MnCO_3$, $FeBO_3$, etc.) and with tetragonal (D_{4h}^{14}) structure (NiF_2). A characteristic property of this type of antiferromagnet as distinguished from easy-axis antiferromagnets is the absence of degeneracy in the spin-wave spectrum branches in zero magnetic field. If the Dzyaloshinskiĭ interaction is disregarded, then at zero magnetic field the energy gap of one of the branches (the low-frequency branch) is governed by the anisotropy in the basal plane and also by the interaction of the magnetic subsystem with other subsystems (for example, with the nuclear or elastic subsystems). As is well known, however, Dzyaloshinskiĭ interaction can exist in the two types of antiferromagnets mentioned above (rhombohedral and tetragonal).[1] While the Dzyaloshinskiĭ interaction does take place, its character differs in these two types of material. Furthermore, there is a difference in the manner in which the corresponding energy appears in the thermodynamic potential. Thus, in rhombohedral antiferromagnets, where the Dzyaloshinskiĭ interaction is isotropic with respect to arbitrary rotations about the principal crystal axis, and where anisotropy in the basal plane is negligible,[2] the low-frequency AFMR branch is independent of the magnetic field direction in the basal plane starting with relatively weak fields (0.5 to 2 kOe). Furthermore, the energy gap is small at h = 0 and is determined essentially by the magnetoelastic[3] and hyperfine[4] interactions. In the case of NiF_2 the Dzyaloshinskiĭ interaction substantially modifies the spectrum and for the low-frequency AFMR branch effects both a significant energy gap and also a dependence of the branch frequency on the orientation of the magnetic field in the basal plane.

The low-frequency AFMR branch in NiF_2 was first observed experimentally by Richards[5] by means of a submillimeter Fourier spectrometer at T = 4.2 K in magnetic fields up to 50 kOe and at a single fixed magnetic field orientation relative to the crystallographic axes (the magnetic field was directed in the basal plane at an angle of $\sim 12°$ to the [100] axis). As was pointed out by Richards[5], at wavelengths greater than 2.5 mm (corresponding to a resonant field of 2.5 kOe), the sensitivity of the magnetic-field orientation decreased sharply. Thus, in view of the nonlinear dependence of the AFMR frequency on the magnetic field, it was not possible to carry out measurements in fields less than 2.5 kOe and to determine the precise value of the energy gap at H = 0.

The present work on the low-frequency AFMR branch in NiF_2 was undertaken with the goal of a precise determination of the energy gap and also a study of the dependence of the frequency on the magnitude and orientation of a magnetic field lying in the basal plane, in the range from 0 to 130 kOe.

EXPERIMENTAL RESULTS

The low-frequency AFMR branch in NiF_2 was investigated at T = 4.2 K at wavelengths from 0.6 to 3 mm by means of direct-amplification spectrometers in which backward-wave tubes were used as the sources of electromagnetic radiation.[6] Single crystals of NiF_2 were oriented with an x-ray goniometer accurate to 2–3°.

Investigations were carried out at the Institute of Physical Problems, USSR Academy of Sciences, in fields of up to 25 kOe obtained by means of the superconducting solenoid described in[7]. The nickel fluoride single crystals used were grown by S. V. Petrov at the same institute. For fields from 10–130 kOe, the investigation was carried out at the P. N. Lebedev Physics Institute, with the "Solenoid" apparatus N[8], and for fields from 0–10 kOe with a laboratory electromagnet. These measurements were carried out on single crystals grown by P. P. Syrnikov at the A. F. Ioffe Physicotechnical Institute.

The experiments were performed at fixed frequencies. The resonant absorption line was recorded with a continuously varying magnetic field by means of an x-y recor-

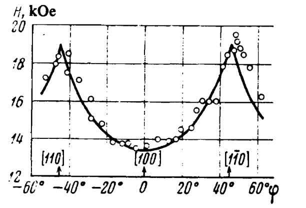

FIG. 1

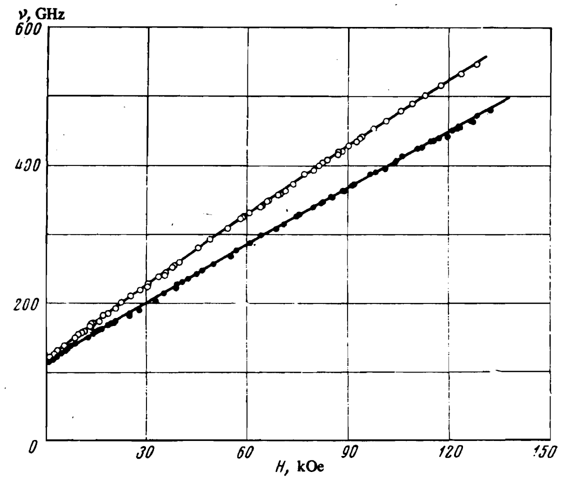

FIG. 2. Dependence of the AFMR frequency on the magnetic field at T = 4.2°K. The open circles are for H∥ [100], and the solid circles are for H∥ [110].

der. The experiments were carried out at a temperature of 4.2 K. Detailed description of the apparatus design and methods of measurement can be found in [9].

Investigations showed that rotation of the magnetic field in the basal plane produces a periodic displacement of the AFMR line with a period of 90° (Fig. 1). The resonant field H_r has a strong angular dependence near the [110] direction and a rather smooth dependence near [100]. It turned out that at frequencies less than 170 GHz the experimental results satisfy the relaxation solid curve in Fig. 1)

$$H_r(\varphi)=H_{100}/\cos\varphi \quad (0\leqslant\varphi\leqslant 45), \tag{1}$$

where H_{100} is the resonant field at H ∥ [100]. At higher frequencies the $H_r(\varphi)$ dependence deviates from (1). For example, the ratio of H_{110} to H_{100} is 1.41 ± 0.03 at 170 GHz as against 1.3 ± 0.03 at 185 GHz.

The ν(H) dependence for fields along [100] and [110] was studied over all the frequency intervals (Fig. 2). A least-squares analysis of the experimental results was carried out by computer. We have obtained a good fit to the data by a polynomial in the form

$$\nu^2=AH^2+BH+C \tag{2}$$

with the following values for the constants: for **H** ∥ [100]

$$A=10.32\pm0.77\ \mathrm{GHz^2/kOe^2},\quad B=935\pm10\ \mathrm{GHz^2/kOe},\quad C=12700\pm200\ \mathrm{GHz^2} \tag{3}$$

and for **H** ∥ [110]

$$A=7.1\pm0.1\ \mathrm{GHz^2/kOe^2},\quad B=700\pm10\ \mathrm{GHz^2/kOe},\quad C=12400\pm200\ \mathrm{GHz^2} \tag{4}$$

The value ν_0 = 112 ± 1 GHz for the low-frequency AFMR energy gap for all the single crystals studied in our work differs from the value ν_0 = 100 ± 1.5 GHz cited by Richards[5] In fields stronger than 30 kOe the Richards data lie close to our experimental curve.

DISCUSSION OF THE RESULTS

There are several possible theoretical interpretations of the data for the low-frequency AFMR branch in NiF_2.[10-13] Interpretation in terms of the thermodynamic potential in the form assumed by Dzyaloshinskiĭ is complicated because of the impossibility of using the Landau-Lifshitz equations to describe the dynamic properties of antiferromagnetic nickel fluoride, for which the standard spin-wave theory assumption

$$\mathbf{LM}=0,\quad \mathbf{L}^2+\mathbf{M}^2=\mathrm{const}, \tag{5}$$

is not satisfied. Here $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$, $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$, and $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations.

A possible means of describing the dynamic effects is afforded by the Turov model[10], where it is assumed that (5) is satisfied by the sublattice mechanical moments, which are related to the sublattice magnetizations by the anisotropic g-factor. The low-frequency AFMR branch for **H** ∥ [100] is given[10] within this model by $(\nu/\gamma)^2 = H^2 + (5H_D - 4H_\tau)H + H_{AE}^2 + 4(H_D - H_\tau)^2$, where γ is the gyromagnetic ratio, and H_{AE}, H_D, and H_τ are the effective fields corresponding to anisotropy in the basal plane and to the transverse and longitudinal Dzyaloshinskiĭ interactions. Four undetermined quantities appear in this equation, whereas the analysis of our data yields only three relations connecting these constants. Thus the resonance measurements alone are insufficient to extract all of the undetermined constants and make a correct comparison of the resonance data with static measurements. Use of the results of static measurements for the analysis of the AFMR data by means of the Turov theory does not seem possible. Actually, within the Turov theory the parallel (along L) magnetic susceptibility is $\chi_\parallel = 0$, whereas it has been shown experimentally[13] that $\chi_\parallel \neq 0$. Detailed discussion of several theoretical explanations of the static and dynamic properties of tetragonal antiferromagnets of the NiF_2 type can be found in[12,13][1)]

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[1)]In [12,13] the Richards value [5] for the low-frequency AFMR branch energy gap was used to calculate the effective longitudinal Dzyaloshinskiĭ-interaction field $H_D^\parallel$. Calculation of $H_D^\parallel$ by A. N. Bazhan using the va?ue of the energy gap obtained in our work has shown that the value for $H_D^\parallel$ agrees within experimental error with that presented in [12,13].

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