

# Doubling of the sound frequency and acoustic detection in hematite

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The first of many possible manifestations of effective anharmonicity (of magnetoelastic origin) of the elastic subsystem of antiferromagnets—second-harmonic generation and acoustic detection of a modulated sound wave—have been observed for the first time in single-crystal hematite. In agreement with the predictions of the theory, both effects are large and depend strongly on the magnetic field.

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Nonlinear acoustic phenomena in solids are potentially diverse and contain much information. Their investigation is made difficult by the relatively low elastic moduli of third order  $\hat{C}^{(3)}$ , although it is possible at sufficiently large sound-wave amplitudes.<sup>[1]</sup> In magnetically ordered crystals, the moduli  $C^{(3)}$  for low-frequency quasielastic oscillations are renormalized because of the nonlinearities of the magnetoelastic (ME) interaction in the magnetic subsystem itself. In weakly anisotropic antiferromagnets (AF) the expected renormalized<sup>[2]</sup> is particularly large because the AF are characterized by participation of exchange interaction in the long-wave oscillations,<sup>[3–5]</sup> and is of particular interest because of its strong dependence on the magnetic field.

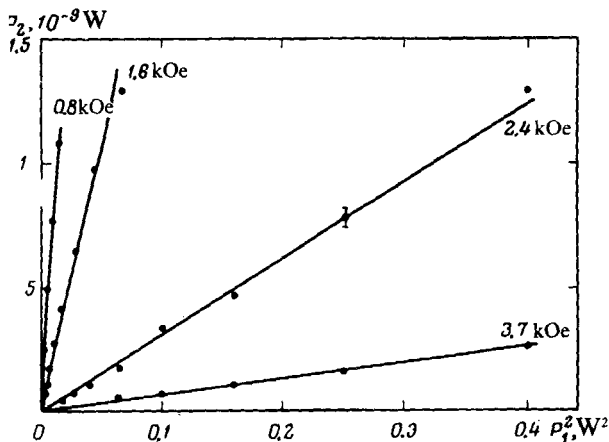


FIG. 1. Second-harmonic signal power vs. the square of the fundamental-harmonic power at different values of the constant magnetic field  $H$ , oriented perpendicular to  $C_1$  and  $U_2$ .

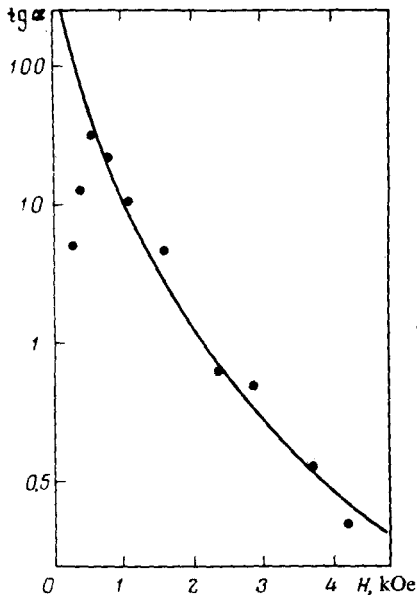


FIG. 2. Field dependence of the efficiency of sound conversion into the second harmonic. Solid curve—result of calculation by formula (27) of [2] matched to the experimental data obtained in a field 2.4 kOe.

The most convenient for an experimental verification of the hypothesis advanced in,<sup>[2]</sup> that the elastic AF subsystem is effectively anharmonic, is hematite ( $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>, near point  $T_N = 960$  K, exchange field  $H_E = 9.2$  MOe) in the easy-plane phase  $T_N > T > T_M = 262$  K, all the more since the larger renormalization, which is independent of the ME field, of the second-order moduli  $\hat{C}^{(2)}$  was confirmed experimentally in this AF.<sup>[6,7]</sup>

Our measurements were performed at room temperature on a synthetic hematite single crystal in the form of a plate of area 0.4 cm<sup>2</sup> and thickness 0.3 cm; the axes  $C_3$  and  $U_2$  were in the plane of the plate. We used a pulse procedure ( $\tau = 1 \mu\text{sec}$ ), wherein the sound was excited with a quartz converter at 34 MHz and was received at double the frequency by another quartz converter. The acoustic contact between the converters and the sample was with the aid of salol. The results were essentially the same for different directions of the sound polarization in the plane of the plate. We present below data on the polarization of the oscillations of the input and output converters along  $C_3$  and  $U_2$ , respectively<sup>1)</sup>. The constant magnetic field  $\mathbf{H}$  was directed perpendicular to the plane of the plate.

A series of second-harmonic pulses was observed, whose amplitude depended strongly on the value of  $H$ . Substitution of fused quartz for the hematite and inclusion of a second-harmonic filter in front of the sample has shown that the second harmonic is not produced in the system itself. We measured the dependences of the amplitude of a number of peaks of the series on the fundamental-frequency signal power at different values of  $H$ . The input RF power was varied with a calibrated attenuator in the range  $10^{-2}$ –1 W, corresponding to strains  $\epsilon_1 \approx (0.4\text{--}4) \times 10^{-7}$  in the primary sound wave. Figure 1 shows plots of the power  $P_2$  of the second-harmonic signal against  $P$  at several values of  $H$ . It is seen that they take the form of straight lines with different

slopes. The slope (tangent of the angle  $\alpha$ ) of these lines, which characterizes the efficiency of conversion of the sound into the second harmonic, is plotted on a logarithmic scale in Fig. 2 as a function of  $H$ .

Thus, the second-harmonic power of the sound has a characteristic quadratic dependence on the fundamental-signal power. The magnetic origin of the effect is confirmed by its strong dependence on  $H$  (Fig. 2). The presence of a maximum on the experimental plot can be easily attributed to the increased attenuation of the sound in the weak-field region, where the magnetoelastic interaction and the influence of the domains are very large. In the field region  $H > 0.8$  kOe, there is good agreement with the theoretical proportionality to  $\omega_0^{-4}$ , where  $\omega_0$  is the antiferromagnetic-resonance frequency.<sup>[2]</sup> This dependence is shown solid in Fig. 2. Under the conditions of our experiment, the efficiency of the conversion of the sound power into the second harmonic amounts to  $\sim 1\%$  in a field of 1 kOe and at strains  $\epsilon_1 \approx 4 \times 10^{-7}$  in the primary wave, which is likewise in agreement with the conclusions<sup>[2]</sup> and is larger by two orders of magnitude than in purely elastic crystals.<sup>[1]</sup>

Besides second-harmonic generation, we observed an effect with a strong dependence on the magnetic field, namely acoustic detection of a modulated sound wave. Sound of 34 MHz frequency was meander-modulated and the signal was received at the meander frequency 390 kHz by a resonant piezoceramic converter. It turned out that the power of the detected signal depends quadratically on the power of the fundamental signal, and its dependence on  $H$  was of the same character as for the frequency doubling (Fig. 2).

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<sup>1</sup>It should be noted that in accordance with Eqs. (16) and (B.1) of<sup>[2]</sup> the doubling effect can be observed in the presence of at least two components of the wave vector of the excited sound. In our case this condition was satisfied because of the multiple reflections of the sound from the sample boundaries (including the side boundaries).

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