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Inversion ratio in trivalent-iron doped andalusite in the Q and V-bands

V. V. Mishenko and N. T. Cherpak

Institute of Radiophysics and Electronics, Academy of Sciences of the Ukrainian SSR, Kharkov-85, USSR

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Résumé. — Les valeurs mesurées de l'indice d'inversion I de l'andalousite (Al₂SiO₅: Fe³⁺) à $\theta = 90^{\circ}$ aux fréquences 35-55 GHz se trouvent dans l'intervalle 1,6-0,6. La dépendance en fréquence de I montre, que la cross-relaxation harmonique détériore essentiellement les propriétés du maser à andalousite près de la fréquence 36 GHz et rend impossible le travail du maser à cette orientation dans l'intervalle de fréquences 47-53 GHz.

Abstract. — Measured values of the inversion ratio I in and alusite (Al₂SiO₅: Fe³⁺) at $\theta = 90^{\circ}$ within the frequency range 35-55 GHz are in the range 1.6-0.6. The frequency dependence of I indicates that the harmonic cross-relaxation markedly reduces the maser properties of and alusite near the frequency 36 GHz. This effect leads to a decrease of I such that maser operation is impossible for range 47-53 GHz at this orientation.

Among the known maser crystals, and alusite $(Al_2O_3.SiO_2)$ with Fe^{3+} ions has the highest value of zero magnetic field splitting. This advantage of and alusite is especially important for solid-state masers of the millimeter (mm) wave band [1]. However, in order to develop a mm maser, it is necessary to know the frequency dependence of the inversion ratio I. In cases where cross-relaxation processes are absent, this parameter is determined by the ratio of the signal frequency and the pump frequency and that of the spin-lattice relaxation transition probabilities. Cross-relaxation may affect the value of the inversion ratio. Unfortunately, it is impossible to calculate probabilities of either the spin-lattice relaxation (w_{cr}) in the proper way.

At frequencies below 100 GHz, the three-level inversion scheme seems to be the most reasonable, realizable with an external magnetic field applied along the optical axis of the crystal ($H \parallel c$ and perpendicular to the magnetic Z axis). With this orientation, it is possible to use both andalusite magnetic complexes in the simplest way. However, analysis [2] of the ESR spectrum indicates that it is impossible to obtain inversion of energy level populations in the frequency range 75-100 GHz because of the two-spin cross-relaxation (CR), since $v_s \simeq \frac{1}{2} v_p$. v_s and v_p are the signal and pump frequency, respectively, $v_s = v_{12}$, $v_p = v_{13}$ or v_{14} . It is interesting to note that in the mm band, multi-spin cross-relaxation (MCR) processes may take place. To find out the influence of these MCR processes on the inversion ratio, it is necessary to know its frequency dependence in a wide frequency

range. There are no sufficient experimental data [2, 3] to determine the frequency dependence of I.

In this paper, we present results of measuring the inversion ratio in andalusite within the frequency range 35-55 GHz. All the experiments were made using natural samples $0.5 \times 0.8 \times 15 \,\mathrm{mm}^3$ in size mounted in a rectangular waveguide of a $5.2 \times 0.8 \,\mathrm{mm}^2$ cross-section as shown in figure 1 and in slow-wave comb structure [4]. The Fe³⁺ concentration in the sample was $0.07 \,\mathrm{wt}$ %. Using the waveguide method for measurements of *I*, we can carry out experiments over a wide range of signal frequencies.

The diffraction radiation generator was used as a pumping source providing output power not less than 1 W in the whole pumping frequency range (132-160 GHz) [5]. The pumpwaves were simply injected into the signal waveguide through pump waveguides. The magnetic field was produced by a superconducting magnet with ferromagnetic screens.

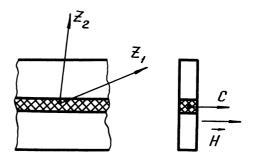


Fig. 1. — The waveguide section with an experimental sample of and alusite crystal; Z_1 and Z_2 are magnetic Z axes.

The field homogeneity is good enough not to broaden noticeably the ESP line. The measurements were carried out at a temperature of T = 1.8 K.

The value of the maximum inversion ratio at different frequencies was determined from its dependence on the pumping power. Such a procedure allowed us to exclude measurement errors arising from the possible weak saturation of the pump transition. In most cases, the value of I was 0.90 to 0.95 of the maximum accessible one.

The dependence of the maximum accessible values of *I* on the signal frequency is given in figure 2. Near the frequencies 36 GHz and 50 GHz, the value of the maximum accessible inversion ratio decreases sharply.

These frequencies are in good agreement with the calculated one at which the conditions

$$2 v_{12} = v_{23} \simeq v_{24}$$
 and $3 v_{12} = v_{23} \simeq v_{24}$

hold. Hence, we can state that at the frequency of 36 GHz, I decreases due to a four-spin harmonic cross-relaxation (HCR), while at 50 GHz because of three-spin HCR. The decrease of the inversion ratio depends on the number n of spins involved in the elementary act of HCR and conforms to the physical notions on the reduction of its influence upon I with growing n.

The frequency dependence of I at $\theta = 90^{\circ}$ indicates that the four-spin HCR, although markedly reducing the maser parameters near 36 GHz, still leaves it operable. Meanwhile, the three-spin HCR leads to a decrease of I such that it is impossible to use this orientation of andalusite for the frequency range 47-53 GHz.

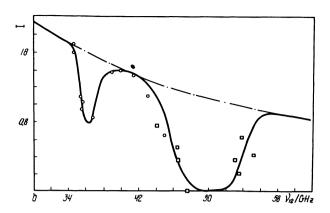


Fig. 2. — The frequency dependence of the inversion ratio I for Al_2O_3 , Fe_2O_3 , Fe^{3+} . a) — .— .— calculated when the cross-relaxation processes are absent; b) —— calculated using the experimental determined w_{cr} . \bigcirc and \square are the experimental points for waveguide section and slow-wave comb structure, respectively.

According to the measurements of I and the physical ideas concerning the HCR probabilities at different numbers of n, one may conclude that a five-spin HCR should not influence I to a noticeable degree. Thus, at frequencies below 30 GHz, I can be described by a smooth function. At the same time, the two-spin HCR would influence it very strongly in a wide range near 100 GHz. As we see, measurements of the inversion ratio with a transverse orientation of andalusite in the frequency range 34-55 GHz has allowed the determination of frequency regions where this inversion scheme is not efficient for masers.

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