# Supplemental Material to the article

# “Magnetic irreversibilities and nonreciprocity of the microwave absorption in $\mathrm{FeCr_2O_4}$ spinel”

**1. Energy level scheme of the $\mathbf{Fe^{2+}}$ ions at $\boldsymbol{T < T_{OO}}$.** As it was explained in [1], the effective Hamiltonian of the $\mathrm{Fe}^{2+}(^5E_\vartheta)$ ions has the form

$$H = \mu_B \mathbf{H}_{\mathrm{mol}}\mathbf{S} + D(3S_z^2 - S(S+1)) + \mu_B\left(g_e - \frac{6\lambda}{\Delta}\right)\mathbf{BS} + \frac{6\lambda\mu_B}{\Delta}S_zB_z, \tag{S1}$$

where $H_{\mathrm{mol}}$ – an average exchange (molecular) field in the ferrimagnetic phase, $D = -\lambda^2/\Delta + \rho$ – magnetic anisotropy of the easy plane type originating from the joint action of the cooperative Jahn–Teller effect and spin-orbit coupling [2], and the last two describe the interaction with a magnetic field. The result of the numerical diagonalization of the operator (S1) when choosing the spin quantization axis along the direction of magnetization is presented in Table S1.

**Table S1.** Energies and wavefunctions of the $\mathrm{Fe}^{2+}$ ion ground state. Parameter values of Eq. (S1) Hamiltonian are $D = 3.07\,\mathrm{cm}^{-1}$, $\lambda/\Delta = -0.03$, $\mu_B H_{\mathrm{mol}} \sim 60\,\mathrm{cm}^{-1}$

| $\vert M_s = 2\rangle$ | $\vert M_s = 1\rangle$ | $\vert M_s = 0\rangle$ | $\vert M_s = -1\rangle$ | $\vert M_s = -2\rangle$ | $E$, $\mathrm{cm}^{-1}$ |
|---|---|---|---|---|---|
| 0.004 | 0 | −0.080 | 0 | 0.997 | 0 |
| 0 | 0.111 | 0 | −0.994 | 0 | 74 |
| −0.107 | 0 | 0.991 | 0 | 0.080 | 141 |
| 0 | 0.994 | 0 | 0.111 | 0 | 199 |
| 0.994 | 0 | 0.107 | 0 | 0.005 | 246 |

From Table S1 we see that the upper estimate of the magnetic moment of the $\mathrm{Fe}^{2+}$ ion if it lies in the $(ab)$ plane of the crystal is $4.39\,\mu_B$. Assuming that the spins of the chromium ions also lie in the $(ab)$ plane, we obtain an estimate of the total magnetic moment of $1.61\,\mu_B$ per formula unit. This value is closer to the value of $\sim 1.1\,\mu_B$ observed in the experiment, but still exceeds it markedly. Therefore, it is logical to assume that the spins of the chromium ion sublattice are noncollinear.

**2. Interference of the magnetic and electric dipole transitions.** The ferromagnetic resonance mainly involves the spin subsystems of chromium and iron ions. The stationary state of the iron sublattice can be described by the Hamiltonian (S1), and the oscillations of the sublattice magnetization under the incident radiation can be considered as small perturbations of these states.

As can be seen from Fig. 4 of the main article, the differences are observed in the absorption intensity in the FMR signal when the sample is rotated 180 degrees. In spite of the fact that collective absorptions are probed, the interaction of individual spins with external electromagnetic radiation plays a fundamental role. The relationship between spins and the electric field in $\mathrm{FeCr_2O_4}$ was discussed in [1]. It was found that in the basis of the states of the $\mathrm{Fe}^{2+}$ ion $(^5E_\vartheta)$ it is described by the effective operator

$$H_E = \frac{4}{3}\sqrt{\frac{7}{2}}d^{(12)3}\frac{\lambda^2}{\Delta}E_z^\omega(S_xS_y + S_yS_x), \tag{S2}$$

where the coupling parameter $d^{(12)3} \cong -0.5\,\mathrm{a.u./e}$ [1].

The probability of the transition is defined by the sum

$$W \propto |\langle f|H_E + H_M|i\rangle|^2 = |\langle f|H_E|i\rangle|^2 + |\langle f|H_E|i\rangle|^2 - \langle f|H_E|i\rangle\langle i|H_M|f\rangle - \langle f|H_M|i\rangle\langle i|H_E|f\rangle, \tag{S3}$$

where $H_M = n\mu_B g\mathbf{S}\mathbf{H}^\omega$ is the interaction of the magnetic moment with the electromagnetic wave field, and $n$ is the refractive index. The last two terms in Eq. (S3) describe the interference effect. As can be seen from Table S1, approximate eigenfunctions and eigenvalues in the frame have the form

$$\begin{gathered} |M_s\rangle, \\ E_{M_s} = |\mathbf{H}_{\mathrm{mol}} + \mu_B g_{\mathrm{eff}}\,\mathbf{B}|M_s. \end{gathered} \tag{S4}$$

The transition matrix element between the ground state and the nearest excited state is

$$\mathrm{Im}\langle -1|H_E + H_M| - 2\rangle = \left\{ \begin{array}{c} \sqrt{\frac{7}{2}}\,\frac{4d^{(12)3}\lambda^2}{\delta}\,\frac{E_c^\omega (M_a^{(\mathrm{Fe})^2} - M_b^{(\mathrm{Fe})^2})}{M^{(\mathrm{Fe})}M^{(\mathrm{Fe})}} \\ + \frac{ng\mu_B E_c^\omega (M_a^{(\mathrm{Fe})}k_a^\omega + M_b^{(\mathrm{Fe})}k_b^\omega)}{|E_c^\omega| M^{(\mathrm{Fe})}} \end{array} \right\}. \tag{S5}$$

We see that the interference terms included in the transition probability are proportional to

$$|E_c^\omega|(M_a^{(\mathrm{Fe})^2} - M_b^{(\mathrm{Fe})^2})(M_a^{(\mathrm{Fe})}k_a^\omega + M_b^{(\mathrm{Fe})}k_b^\omega). \tag{S6}$$

It follows from this expression that the effect of non-reciprocity is evident in the configuration of the fields shown in Fig. S1.

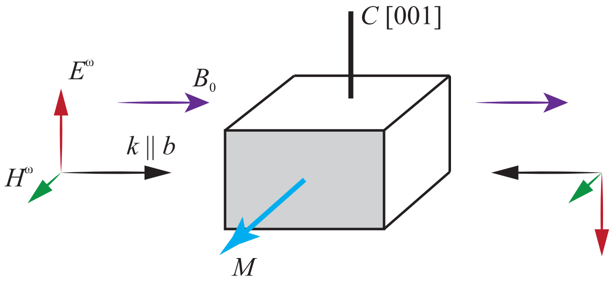

Fig. S1. Model configuration for Eq. (S1)

Estimates of transition probabilities are shown in the diagram in Fig. S2 at an external field value of 800 mT.

Fig. S2. Transition probability diagram in relative units. The transition from the ground to the nearest excited spin level of the $\mathrm{Fe}^{2+}$ ions is considered

A more accurate numerical diagonalization of the operator Eq. (S1) results in the diagram shown in Fig. S3, which qualitatively corresponds to the obtained experimental results.

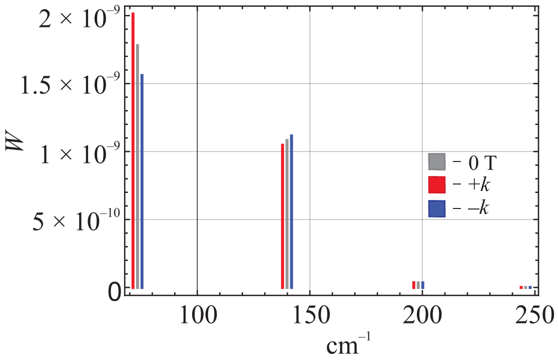

Fig. S3. Absorption probability diagram showing the effect of nonreciprocity due to the interference of magnetic and induced electric dipole transitions in a magnetic field of 800 mT

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1. K. V. Vasin and M. V. Eremin, J. Phys.: Condens. Matter **33**, 225501 (2021).
2. K. V. Vasin and M. V. Eremin, J. Magn. Magn. Mater. **537**, 168185 (2021).