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THz Magneto-electric atomic rotations in the chiral compound $Ba_3NbFe_3Si_2O_{14}$.

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THz absorption spectra in the two $Ba_3NbFe_3Si_2O_{14}$ crystals

Figure 1 presents the absorption/absorbance spectra (left/right scales) measured with the 6 μm BMS on two different samples. The absolute absorbances, αd, with α the absorption and d the travelled distance through the sample, were determined by measuring the transmission through a 2 mm diaphragm as a reference and the sample transmission through that same diaphragm. Interference pattern from the sample surfaces is present for sample 2 (lower panel). For further analysis, this interference pattern was removed using a band block FT filter. The larger oscillations observed below 30 cm^{-1} for sample 1 (upper panel) are instrumental. For further analysis, they were removed using the sample at a different temperature as a reference. The stronger absorption observed for **e//c** is attributed to a (multi-)phonon band.

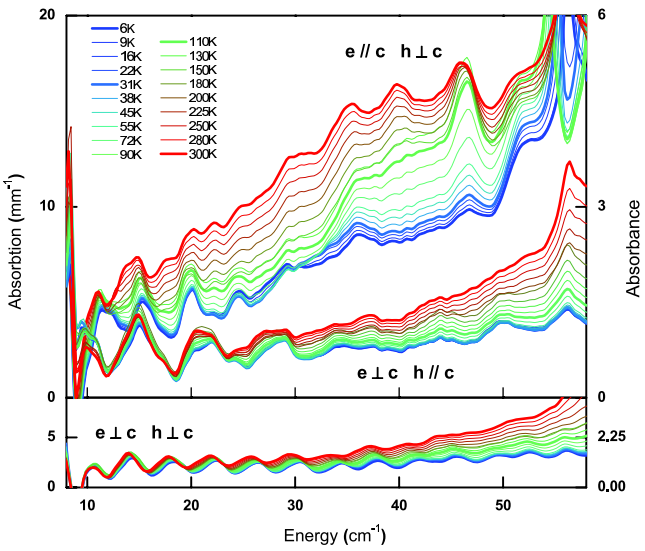

FIG. 1: **Absorption spectra in $Ba_3NbFe_3Si_2O_{14}$ measured with the 6 μm BMS**. The temperature was varied from 6 K to 300 K. Three different polarizations of the THz electromagnetic field as regards the crystal **c**-axis were measured. Upper panel: measurements on sample 1, (**e//c h⊥c**) and (**e⊥c h//c**). Lower panel: measurements on sample 2 (**e⊥c h⊥c**).

Interaction between R_z and the external fields in Eq.1

In isotropic or cubic systems, the external field conjugated to rotations is a torque. However, in systems with reduced symmetry, the linear coupling between rotations $\mathbf{R}$ and linear displacements can be allowed. This is precisely our case, since our system initially has D_3 point-group symmetry which is further reduced to C_2. This means that $\mathbf{R}$ in our system is not a pure rotation anymore, so that the standard forces acting on the above displacements also act on $\mathbf{R}$. Polar displacements implying a relative motion of charges are among the displacements linearly coupled to $\mathbf{R}$. Thus, the electromagnetic radiation is coupled to rotations via this motion of charges, which gives rise to the unusual electromagnetic activity of the atomic rotations in our system.

More specifically, the charges feel the electromagnetic radiation through the Lorentz force: $\mathbf{f} = -\frac{e}{c}\frac{\partial \mathbf{A}}{\partial t} - e\nabla\phi + \frac{e}{c}\dot{\mathbf{r}} \times (\nabla \times \mathbf{A})$, where $\mathbf{r}$ represents the position of the charge e, while ϕ and $\mathbf{A}$ are the scalar and vector potentials of the radiation respectively [1]. In the Weyl gauge ($\phi = 0$), the vector potential of a monochromatic plane wave is $\mathbf{A}(\mathbf{r}, t) = \mathbf{a}_0 e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} \simeq (1 + i\mathbf{k}\cdot\mathbf{r})\mathbf{A}_0(t)$ in the long-wavelength limit. Here $k = n\omega/c$ and $\mathbf{A}_0(t) = \mathbf{a}_0 e^{-i\omega t}$, with n being the refractive index of the medium and $\mathbf{a}_0$ a constant complex vector. In this limit, the first term in the vector potential generates the electric field of the wave $\mathbf{E} = i(k/n)\mathbf{A}_0$, while the magnetic field $\mathbf{H} = i\mathbf{k} \times \mathbf{A}_0$ is obtained from the second one. Keeping this in mind, the vector potential can be rewritten as $\mathbf{A} \simeq \frac{n}{ik}\mathbf{E} + \mathbf{H} \times \mathbf{r} + n\frac{\mathbf{k}}{k}(\mathbf{E}\cdot\mathbf{r})$. Accordingly, the Lorentz force becomes $\mathbf{f} \simeq e\mathbf{E} + \frac{e}{c}\mathbf{r} \times \dot{\mathbf{H}} + ie\mathbf{k}(\mathbf{E}\cdot\mathbf{r}) + \frac{e}{c}\dot{\mathbf{r}} \times \mathbf{H}$. We note that the second and the last contribution to this force are sort of dual contributions. Interestingly, the last term was invoked by Dzyaloshinskii and Mills in order to explain the unexpected paramagnetic behavior observed in nonmagnetic ferroelectrics [2]. In our case, the second term is the key one for the physical interpretation of the absorption spectrum of our system. In fact, in our measurements, the last two contributions to the Lorentz force produce subdominant effects and therefore can be ignored. Furthermore, this force acts directly on the charge distribution of the system, and therefore on its electric polarization $\mathbf{P} = \sum e_i \mathbf{r}_i$. But since this polarization is linearly coupled to atomic rotations in our system, this gives rise to an effective force acting on the rotations as explained above. The force acting the rotations R_z about the z-axis, in particular, can be written as

$$f_{\text{eff}} = \frac{\alpha}{\omega_{P_z}^2} E_z + g\frac{\alpha}{\omega_{P_z}^2}(\mathbf{P}_\perp \times \dot{\mathbf{H}}_\perp)_z. \tag{1}$$

Here α represents the strength of the coupling between R_z and P_z, ω_{P_z} is the frequency (or stiffness) associated to P_z, and g is a gyromagnetic factor. This force can be obtained as $f_{\text{eff}} = \frac{\delta L_{\text{int}}}{\delta R_z}$, where L_{int} is the Lagrangian in Eq. (1) of the main text.

If $\mathbf{P}_\perp$ is uniform, both electric and magnetic-field components of the THz radiation will couple to $q = 0$ rotations due to momentum conservation. However, if $\mathbf{P}_\perp$ is modulated with a finite wavevector Q, then L_{int} becomes

$$L_{\text{int}} = \lim_{k\to 0}\Big[\alpha_1 E_z(k) R_z(-k) + \alpha_2\big(\mathbf{P}_\perp(-Q) \times \dot{\mathbf{H}}_\perp(k)\big)_z R_z(Q-k) + (Q \to -Q)\Big] \tag{2}$$

in Fourier space, where $\alpha_1 = \alpha/\omega_{P_z}^2(0)$ and $\alpha_2 = g\alpha/\omega_{P_z}^2(Q)$. In this case, the electric field couples to $q = 0$ rotations while the magnetic field does to $q = \pm Q$ ones, thus probing different resonant energies.

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