Strong dynamic magnetoelectric coupling in metamaterial

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Abstract. Magnetoelectric susceptibility ($\xi$) of classical magnetoelectrics is always substantially smaller than the geometric average of the electric ($\chi_e$) and magnetic ($\chi_m$) susceptibilities. Materials reaching the equality $\xi^2 = \chi_e \chi_m$ as a limiting value may thus be considered as ideal magnetoelectrics. Here we investigate the magnetoelectric susceptibility of a metamaterial built from split ring resonators both experimentally and within an equivalent circuit model. Due to a direct proportionality between electric polarization and magnetization, the magnetoelectric susceptibility of this metamaterial fulfills the equality $\xi^2 = \chi_e \chi_m$ at least in the dynamic regime.

1 Introduction

Magnetoelectric effect manifests a connection between electricity and magnetism and has been predicted already 1860 by Pierre Curie, but could be confirmed experimentally only about half a century ago [1,2]. Up to now the experimentally observed effects remain rather weak. The weakness of the magnetoelectric susceptibility can to some extent be explained by the absence of strong mechanisms which couple magnetism and electricity on the microscopic level [3]. However, as has been shown theoretically [1,4], the allowed limiting value of the magnetoelectric susceptibility ($\xi$) is quite large and equals to the geometric average of electric ($\chi_e$) and magnetic ($\chi_m$) susceptibilities:

$$\xi^2 \leq \chi_e \chi_m.$$  (1)

In classical magnetoelectric materials like Cr$_2$O$_3$ the limiting value of equation (1) is failed by about two orders of magnitude [2,4]. In efforts to increase the value of the magnetoelectric effect, materials revealing both strong electric and magnetic susceptibilities have been brought into consideration. Especially close to phase transitions, the electric and magnetic susceptibilities may diverge in ferroelectrics and ferromagnets. Materials simultaneously showing the ferroelectricity and ferromagnetism are called multiferroics and they are presently the subject of intensive research [2,5,6].

In case the equality in equation (1) holds, it can be rewritten in the form $\xi = \sqrt{\chi_e \chi_m}$ and the constitutive relationships

$$M = \chi_m H + \chi_{me} E$$
$$P = \chi_e E + \chi_{em} H,$$

(2)
(3)

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estimate for the limiting value of the magnetoelectric effect \( \text{Re}(\xi) \leq \text{Im}(\sqrt{\eta}) \) has been obtained \[22\].

Many designs of metamaterials are based on split ring resonators \[27\]. These elements can be seen as the smallest possible representations of the well known LC-circuit with a single inductance loop as given by the metallic ring and a tiny capacitance produced by the gap in the ring \[28\]. Split ring resonators have been originally developed to achieve negative magnetic permeability which is a key property for design of metamaterials with negative refraction \[8\]. As has been realized recently \[29\], the split ring resonators strongly modify the interactions with electromagnetic radiation by introducing a so called bianisotropy term into the set of basic equations \[30\], which is closely connected to the magnetoelectric effect \[2,6\]. This additional term in the constitutive relations cross-couples the magnetic and electric fields within a split ring resonator \[31,32\]. The bianisotropy offers another degree of freedom \[33\] in controlling the properties of light. Here we note that, contrary to chiral metamaterials, the bianisotropy does not automatically lead to polarization rotation for geometries parallel to the principal optical axes. In order to obtain polarization rotation, these structures must be tilted or measured within off-axis geometry. The corresponding effects \[34,35\] have been termed extrinsic chirality.

In this work we show that metamaterials built from split ring resonators achieve magnetoelectric effects equal to the theoretically limiting value of equation (1). To prove this we investigate a metamaterial of split ring resonators within different geometries, especially including those sensitive to magnetoelectricity. This allowed to obtain electric, magnetic and magnetoelectric susceptibilities and compare them with a simple circuit model. We show that the metamaterial investigated indeed reaches the theoretical limit for magnetoelectric coupling at least in the dynamic regime. This value is due to a direct proportionality of electric and magnetic moments in split ring resonators.

### 2 Experimental details

Transmittance experiments at millimeter-wave frequencies (60 GHz < \( \nu < 120 \) GHz) were carried out in a Mach-Zehnder interferometer arrangement \[36,37\]. This arrangement allows to measure both the intensity and the phase shift of the radiation transmitted through the sample within controlled polarization geometries. The split ring resonator arrays used in present experiments were prepared by chemical etching of copper-laminated board. The rings are typically 0.35 mm × 0.35 mm in size with the gap width \( d = 0.17 \) mm. The lattice constant of the metamaterial is \( l = 0.7 \) mm. The characteristic parameters of various sets of the split ring resonators have been varied within approximately a factor of two and showed qualitatively similar results. Woven glass with a thickness of 0.56 mm was used as a non-conductive substrate. The refractive index of the substrate has been determined in a separate experiment as \( n_s = 2.07 + 0.04i \).

### 3 Results and discussion

Split ring resonators seem to represent a magnetoelectric material fulfilling the condition \( \chi_{me}\chi_{em} = \chi_{e}\chi_{m} \). Indeed, from the effective RLC-circuit model and simple calculations \[29,38\] one can easily get

\[
\chi_e = nCd^2F(\omega); \quad \chi_m = nCS^2\frac{\omega^2}{c^2}F(\omega) \tag{5}
\]

\[
\chi_{em} = -\chi_{me} = nCdS\frac{i\omega}{c}F(\omega). \tag{6}
\]

Here \( n \) is the density of the rings, \( d \) and \( C \) are the effective width and capacitance of the gap, \( S \) is the area of the rings, and \( \omega \) is the angular frequency. We use the Lorentz substitution \( F(\omega) = \omega_0^2/(\omega_0^2 - \omega^2 - i\omega\gamma) \) where \( \omega_0 \) and \( \gamma \) are the resonance frequency and width, respectively. The symmetry of the magnetoelectric coefficients is fulfilled automatically in this model (Eq. (6)).

In order to obtain the electrodynamic parameters of the split ring resonators we have carried out the initial experiments within the geometries suggested in reference \[30\] (shown in the insets to Fig. 1). Three relevant geometries in this case are “magnetic” (\( h \) perpendicular to the plane of the rings), “electric” (\( \epsilon \) parallel to the gap of the rings) and “magnetoelectric” (both excitations are realized simultaneously). In these three geometries and within reasonable approximation the effective refractive indexes basically determine the transmittance close to the resonance and they are given by \( n_e = \sqrt{\varepsilon - \xi^2}/\mu, \quad n_m = \sqrt{\mu - \xi^2}/\varepsilon, \) and \( n_{me} = \sqrt{\varepsilon\mu - \xi^2} \), respectively. Here we neglect the influence of the substrate for simplicity. Although the magnetoelectric susceptibility is included in these equations, the dominating terms for typical parameters of the model are given by \( \sqrt{\varepsilon}, \sqrt{\mu}, \) and \( \sqrt{\varepsilon\mu} \), respectively. In all cases the magnetoelectric susceptibility represents a weaker correction under the square root. This is demonstrated in Figure 1 as we set the magnetoelectric susceptibility to zero, which simply leads to a shift of the resonance frequency. In all series of experiments with varying geometries the influence of the magnetoelectric susceptibility was below the experimental accuracy. This accuracy depends not only on experimental uncertainties, but also on the assumptions of the circuit model, like neglecting of the cross coupling effects, or assumption of infinitely small sizes of the rings compared to the wavelength. On the contrary, electric and magnetic geometries robustly depends on electric \( (\chi_e) \) and magnetic \( (\chi_m) \) susceptibilities. Therefore, both susceptibilities may be reliably determined from the spectra in Figure 1.

The result of the experiments described above may now be extended to obtain the magnetoelectric susceptibility using further geometries of the experiment. In order to get better sensitivity to the magnetoelectric susceptibility, the sample must be measured in the tilt geometry and the signals in parallel and in crossed polarizers have to be compared (see inset in Fig. 2). An example of such tilt geometries has been presented in reference \[32\], where the existence of a cross-coupling terms has been detected.