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Giant spatial-dispersion-induced birefringence in metamaterials

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We demonstrate experimentally giant spatial-dispersion-induced birefringence in metamaterials. The difference between the reflection coefficients for $(1,\bar{1},0)$- and $(0,0,1)$-polarized light reflected from the $[1,1,0]$ surface of a metamaterial reaches 78%. The magnitude of spatial-dispersion-induced birefringence in the transparency windows of the structure reaches $n_{1\bar{1}0} - n_{001} = -0.13$, which is at least three orders of magnitude larger than the typical values reported for natural crystals. Our results elucidate the important role of spatial dispersion effects in a wide class of metamaterials.

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The characterization of metamaterials' electromagnetic properties is a cornerstone problem in materials science. Metamaterials physics [1–7] offers unique opportunities not achievable for natural structures, such as negative refraction [8], subwavelength imaging [9], and cloaking [10]. However, there exists a number of phenomena including spatial-dispersion-induced birefringence that are known for natural crystals [11] but have not yet been studied in metamaterials. In the present Rapid Communication we report the results of an experimental investigation of spatial-dispersion-induced birefringence in a three-dimensional metamaterial.

It is known that cubic crystals composed of isotropic particles are optically isotropic within the local effective medium model validity domain [11]. However, as Lorentz pointed out [12,13], the direction of the wave vector of the wave propagating in a crystal provides a selected direction in space. If the material parameters of the structure depend on the wave vector, i.e., spatial dispersion effects are present, the material would acquire anisotropic properties. Spatial dispersion (or nonlocality) is often treated as a weak effect [14]. However, nonlocal effects turn out to be the intrinsic property of a wide class of metamaterials in contrast with the natural crystals [15–18].

The phenomenon of spatial-dispersion-induced birefringence is usually studied experimentally either in transmission types of experiments or in experiments on reflection from the boundary of the crystal parallel to the $[1,1,0]$ symmetry plane [Fig. 1(a)]. The magnitude of the effect is characterized by the difference of refractive indices for two orthogonal linear polarizations of light propagating along the $(1,1,0)$ direction, $\Delta n = n_{1\bar{1}0} - n_{001}$. The values of Δn were measured [19–22] for crystals such as CuBr, CuI, KI, NaI, CsI, NaCl, BaF_2, CaF_2, etc. The typical reported values of Δn for natural materials do not exceed $|\Delta n_{\max}| \approx 5 \times 10^{-5}$. The measured difference between the reflection coefficients for $(1,\bar{1},0)$ and $(0,0,1)$ polarizations was not greater than 1% for Si and Ge [23].

Isotropic metamaterials [24–30] constitute an important class of artificially structured media. Such metamaterials are usually composed of resonant inclusions. We consider a three-dimensional structure composed of isotropic resonant particles arranged in a cubic lattice, shown in Fig. 1(a). Such isotropic meta-atoms can be implemented as plasmonic particles [31–33], or as high-contrast dielectric [34–38] or magnetodielectric [39,40] particles. Regardless of the nature of the resonance (electric or magnetic), a meta-atom can be described by the Lorentzian-like polarizability $\alpha(\omega)$,

$$\alpha^{-1}(\omega) = \alpha_0^{-1}(\omega) - 2i\,\omega^3/(3c^3), \tag{1}$$

$$\alpha_0(\omega) = \frac{A}{\omega_0 - \omega - i\gamma} + \alpha_b, \tag{2}$$

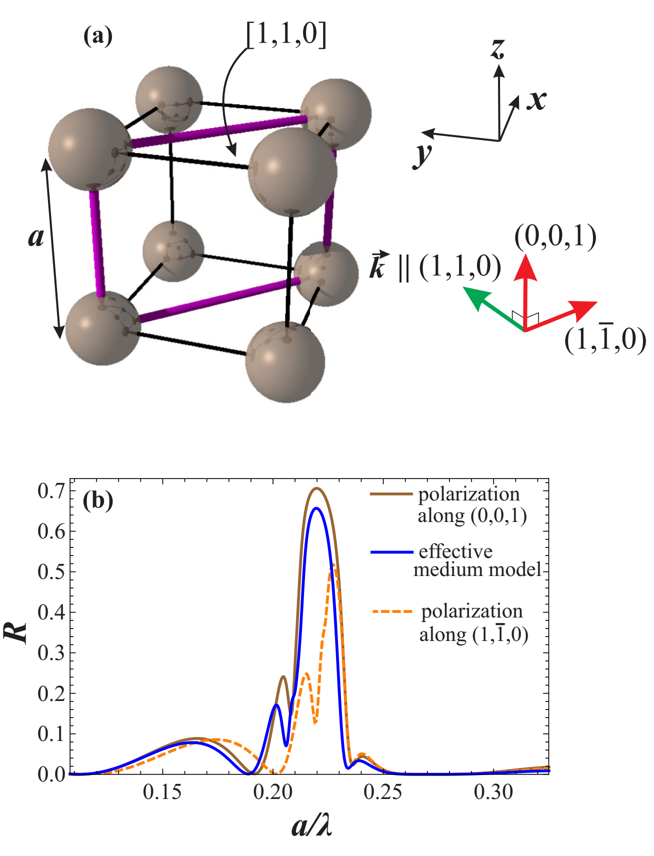

FIG. 1. (a) The view of the three-dimensional metamaterial under study. The boundary of the structure is parallel to the $[1,1,0]$ plane, and the normal incidence of light is studied. The reflection coefficients are measured for $(1,\bar{1},0)$ and $(0,0,1)$ linear polarizations of the impinging wave. (b) Theoretical results for reflectance of the structure with parameters $A/a^3 = 0.15$, $\gamma\,a/c = 1.02 \times 10^{-2}$, $\alpha_b/a^3 = 0.0279$, $\lambda_0/a = 4.5$, where a is the lattice period, $\lambda_0 = 2\pi\,c/\omega_0$, and ω_0 is the particle resonance frequency. Particle polarizability is extracted from numerical simulations in Ansoft HFSS.

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where α_0 is a bare particle polarizability (without a radiation loss contribution), ω_0 is a particle resonance frequency, α_b is a background polarizability due to the substrate, and γ is a term describing losses in the particle material. Typically, the structure in Fig. 1(a) is described by the effective medium model and the effective permittivity (or permeability) of the structure is defined by the Clausius-Mossotti formula [41]

$$\varepsilon^{\rm loc}(\omega)[\mu^{\rm loc}(\omega)] = \frac{1 + 8\pi\,\alpha_0(\omega)/(3a^3)}{1 - 4\pi\,\alpha_0(\omega)/(3a^3)}. \tag{3}$$

The reflection coefficients from the boundary of isotropic material under the normal incidence are the same for all polarizations of the impinging wave. However, a more rigorous theoretical approach based on the discrete dipole model [42–44] reveals a significant difference between the reflectances for (0,0,1) and $(1,\bar{1},0)$ polarizations. The calculated results for a five-layer structure are depicted in Fig. 1(b). Noteworthy, in the vicinity of the individual scatterer resonance when $a/\lambda = 0.22$, the reflection coefficients for two polarizations are calculated to be $R_{001} = 0.70$ and $R_{1\bar{1}0} = 0.30$ and thus the relative difference between them is $\delta = 2\,(R_{001} - R_{1\bar{1}0})/(R_{001} + R_{1\bar{1}0}) \times 100\% = 80\%$. Furthermore, the reflectance for (0,0,1) polarization is well captured by the effective medium model, whereas the reflectance for polarization $(1,\bar{1},0)$ deviates significantly from the effective medium model result. Using the discrete dipole model we also plot refractive indices for both polarizations of the structure eigenmodes (Fig. 2). Our results suggest that in transparency windows of the metamaterial ($\text{Im}\,n/\text{Re}\,n < 0.02$) in the spectral interval $a/\lambda < 0.200$ or $a/\lambda > 0.274$, the anisotropy of the structure can be as large as $\Delta n = -0.13$, which is at least three orders of magnitude larger than that measured for natural cubic crystals. Moreover, there is a drastic difference in the imaginary parts of the refractive indices [Fig. 2(b)], which means that the structure acquires giant dichroism. For example, at the resonance frequency, $\text{Im}\,n_{001}/\text{Im}\,n_{1\bar{1}0} = 14$. On the other hand, losses in the particle material are sufficiently small. Therefore, the large imaginary part of n_{001} in the vicinity of the individual particle resonance should be attributed to the evanescent nature of (0,0,1)-polarized waves in this spectral region. This conclusion is in agreement with the theoretical model [44].

Importantly, the predicted effects associated with spatial dispersion can be manifested at any frequency range from the microwave to the visible domain, depending on the properties of the meta-atoms.

To verify the proposed concept of giant spatial-dispersion-induced birefringence, we fabricated a metamaterial with parameters corresponding to the theoretical ones (Fig. 3). For the sake of fabrication simplicity, we chose a planar realization of meta-atoms that were arranged in the sites of a cubic lattice [Fig. 3(a)]. Note that the reflection coefficients were measured for the normal incidence only in which case polarization perpendicular to the plane of the meta-atom is not excited and the presence of the substrate does not introduce additional anisotropy into the system. Therefore, for this particular situation, the experimental system shown in Fig. 3 perfectly emulates the idealized system in Fig. 1(a).

A planar lattice of resonant isotropic particles [45,46] with the radius $R = 4.8$ mm [Fig. 3(b)] was printed on a common FR4 substrate Kingboard Laminates KB-6164. The period of the manufactured cubic lattice is $a = 16.3$ mm. The typical

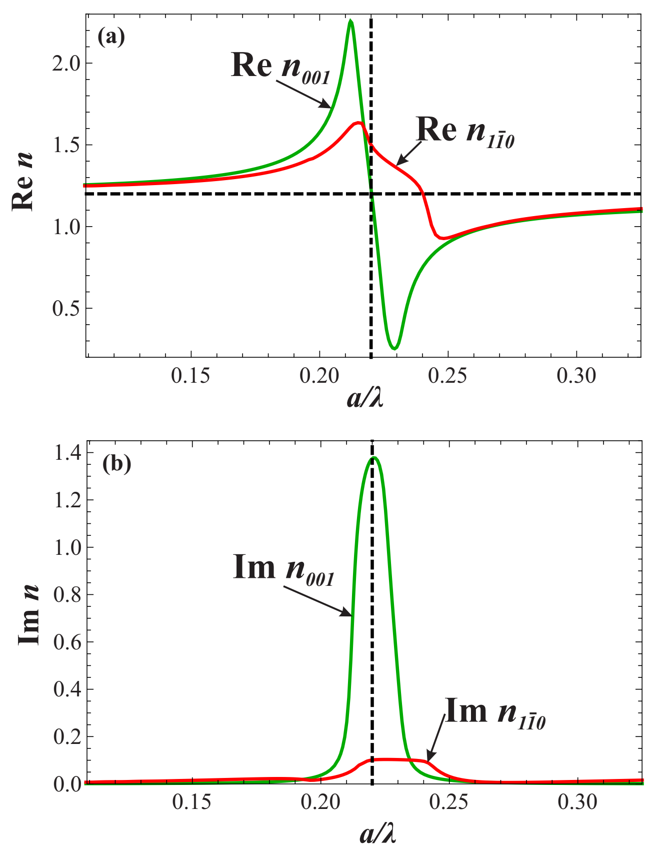

FIG. 2. Effective refractive indices for (0,0,1) and $(1,\bar{1},0)$ linear polarizations of light propagating along the (1,1,0) direction in the metamaterial [Fig. 1(a)]. Calculations are performed by the discrete dipole model. (a) Real part of refractive indices. (b) Imaginary part of refractive indices.

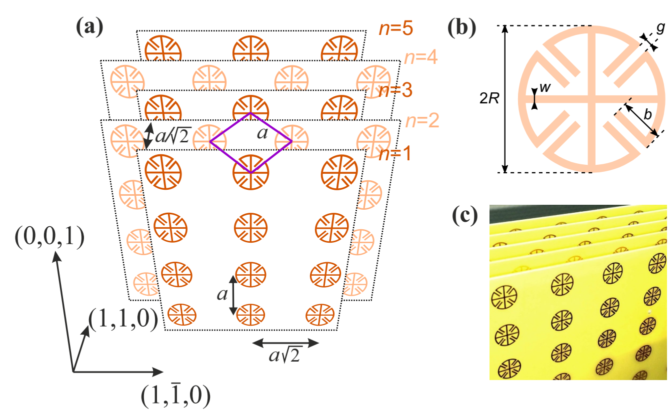

FIG. 3. (a) The sketch of the simplified structure used in the experiment. (b) Resonant copper particles isotropic in plane [1,1,0] having the following geometric parameters: $w = 0.5$ mm, $R = 4.8$ mm, $b = 3$ mm, $g = 0.5$ mm. (c) The photograph of the manufactured five-layer metamaterial sample composed of resonant meta-atoms. The period of the cubic lattice is $a = 16.3$ mm.

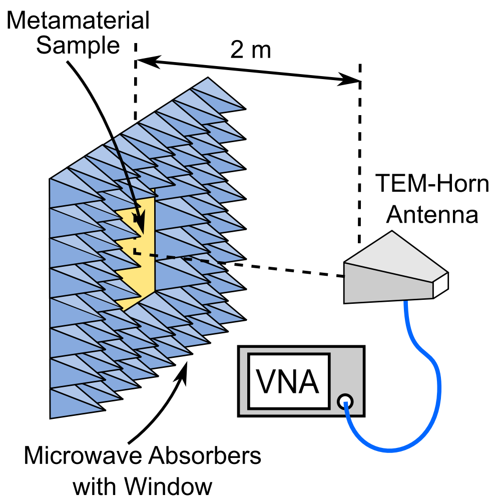

FIG. 4. The scheme of the experiment on the measurement of the reflection coefficients in an anechoic chamber.

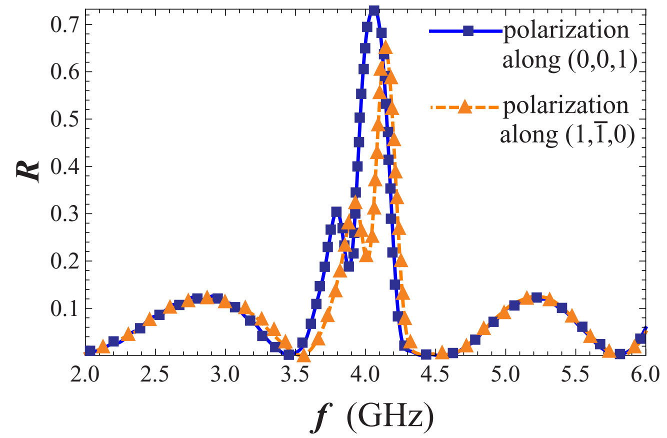

FIG. 5. Experimental results on the reflectance of the five-layer structure composed of resonant meta-atoms.

substrate permittivity in the frequency band under study is $\epsilon_r = 4.58$, the dielectric loss tangent is equal to $\tan\delta = 0.019$, and the thickness of the substrate layer is $h_s = 1.5$ mm. Each layer contains 16×19 particles and has the dimensions 368×309 mm. The bulk metamaterial structure consists of five parallel layers. The assembled five-layer structure was inserted in the window of the same dimensions in a wall covered by microwave absorbers to suppress the influence of the edge diffraction effects at the boundaries of the sample. To measure the reflection coefficients for both orthogonal linear polarizations, the setup with a single TEM-horn antenna was used, as shown in Fig. 4. The horn was installed 2 m away from the sample and connected to a calibrated 50-Ω port of the vector network analyzer Agilent PNA E8362C.

The reflection coefficients were determined in the spectral range from $f_{\min} = 2.0$ GHz to $f_{\max} = 6.0$ GHz. The values of the reflection coefficients were extracted from the measured values of the S_{11} parameter for the metamaterial sample and for the calibrating metal plate of the same dimensions. To suppress the influence of multiple reflections between the studied sample and the horn antenna, the time gating technique was employed [47].

The measured reflectances for two polarizations of the incident wave are depicted in Fig. 5 and they demonstrate a good agreement with the theoretical results for reflectance. In particular, the experimental data show that in the vicinity of the individual particle resonance at frequency 4.03 GHz, the reflectances are $R_{001} = 0.483$ and $R_{1\bar{1}0} = 0.211$ and the relative difference between the reflectances is $\delta = 2(R_{001} - R_{1\bar{1}0})/(R_{001} + R_{1\bar{1}0}) \times 100\% = 78\%$. Noteworthy, the detected anisotropy in reflectance is almost two orders of magnitude larger than that measured for semiconductors such as Si and Ge [23]. Experimental results reveal also that the reflectance maxima for (0,0,1) and (1,$\bar{1}$,0) polarizations correspond to the different frequencies $f_{001} = 4.104$ GHz and $f_{1\bar{1}0} = 4.216$ GHz and thus the frequency shift of the reflectance maxima is $\Delta f = 0.112$ GHz.

With regard to these results it is important to highlight the essentially three-dimensional nature of the structure. For a two-dimensional system [a single layer of particles depicted in Fig. 3(a)], the polarization dependence of the reflection coefficient emerges due to the low symmetry of the lattice even in the absence of spatial dispersion. Therefore, the measurement of reflectances for a single particle layer does not allow one to extract directly the spatial dispersion corrections from the measured reflectance anisotropy. Note that a detailed characterization of the spatial dispersion effects in two-dimensional (2D) structures employing Mueller-matrix spectroscopy and based on the measurements of the reflection coefficients for various incidence angles as well as different azimuthal angles was conducted in Refs. [48,49].

Our experimental results prove the importance of spatial dispersion effects in a wide class of metamaterials composed of resonant particles. It is the resonant nature of inclusions that ensures giant spatial-dispersion-induced birefringence exceeding that in the natural materials at least by three orders of magnitude. The described effects can be observed in any metamaterial consisting of resonant isotropic inclusions (plasmonic or dielectric) arranged in a cubic lattice. Our results suggest that a number of metamaterials deemed to be isotropic in the local effective medium framework can nevertheless acquire pronounced anisotropic properties in a certain spectral range due to spatial dispersion effects.

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[1] S. Linden, C. Enkrich, M. Wegener, J. Zhou, T. Koschny, and C. M. Soukoulis, Science **306**, 1351 (2004).

[2] S. Zhang, W. Fan, N. C. Panoiu, K. J. Malloy, R. M. Osgood, and S. R. J. Brueck, Phys. Rev. Lett. **95**, 137404 (2005).

[3] C. Soukoulis, S. Linden, and M. Wegener, Science **315**, 47 (2007).
[4] V. Shalaev, Nat. Photonics **1**, 41 (2007).
[5] N. I. Landy, S. Sajuyigbe, J. J. Mock, D. R. Smith, and W. J. Padilla, Phys. Rev. Lett. **100**, 207402 (2008).
[6] N. Liu, H. Guo, L. Fu, S. Kaiser, H. Schweizer, and H. Giessen, Nat. Mater. **7**, 31 (2008).
[7] N. I. Zheludev, Science **328**, 582 (2010).
[8] R. A. Shelby, D. R. Smith, and S. Schultz, Science **292**, 77 (2001).
[9] J. B. Pendry, Phys. Rev. Lett. **85**, 3966 (2000).
[10] J. B. Pendry, D. Schurig, and D. R. Smith, Science **312**, 1780 (2006).
[11] V. Agranovich and V. Ginzburg, *Spatial Dispersion in Crystal Optics and the Theory of Excitons* (Wiley-Interscience, New York, 1966).
[12] H. A. Lorentz, Verh. K. Akad. Wet. Amsterdam, Afd. Natuurkd. **18** (1878).
[13] H. A. Lorentz, Proc. R. Acad. Sci. Amsterdam **24**, 333 (1921).
[14] L. Landau and E. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, Oxford, U.K., 1984).
[15] P. A. Belov, R. Marques, S. I. Maslovski, I. S. Nefedov, M. Silveirinha, C. R. Simovski, and S. A. Tretyakov, Phys. Rev. B **67**, 113103 (2003).
[16] C. Menzel, C. Rockstuhl, R. Iliew, F. Lederer, A. Andryieuski, R. Malureanu, and A. V. Lavrinenko, Phys. Rev. B **81**, 195123 (2010).
[17] M. A. Gorlach and P. A. Belov, Phys. Rev. B **92**, 085107 (2015).
[18] S. V. Zhukovsky, A. Andryieuski, O. Takayama, E. Shkondin, R. Malureanu, F. Jensen, and A. V. Lavrinenko, Phys. Rev. Lett. **115**, 177402 (2015).
[19] J. L. Deiss, A. Daunois, A. Chouiyakh, and O. Gogolin, Solid State Commun. **53**, 79 (1985).
[20] F. Meseguer and M. Cardona, Solid State Commun. **50**, 371 (1984).
[21] C. Zaldo, C. Lopez, and F. Meseguer, Phys. Rev. B **33**, 4283 (1986).
[22] J. H. Burnett, Z. H. Levine, and E. L. Shirley, Phys. Rev. B **64**, 241102(R) (2001).
[23] D. E. Aspnes and A. A. Studna, Phys. Rev. Lett. **54**, 1956 (1985).
[24] D. O. Guney, T. Koschny, and C. M. Soukoulis, Opt. Express **18**, 12348 (2010).
[25] T. Koschny, L. Zhang, and C. M. Soukoulis, Phys. Rev. B **71**, 121103(R) (2005).
[26] W. J. Padilla, Opt. Express **15**, 1639 (2007).
[27] J. D. Baena, L. Jelinek, and R. Marques, Phys. Rev. B **76**, 245115 (2007).
[28] A. Grbic and G. V. Eleftheriades, J. Appl. Phys. **98**, 043106 (2005).
[29] P. Alitalo, S. Maslovski, and S. Tretyakov, J. Appl. Phys. **99**, 124910 (2006).
[30] Q. Zhao, B. Du, L. Kang, H. Zhao, Q. Xie, B. Li, X. Zhang, J. Zhou, L. Li, and Y. Meng, Appl. Phys. Lett. **92**, 051106 (2008).
[31] N. G. Khlebtsov and L. A. Dykman, J. Quant. Spectrosc. Radiat. Transfer **111**, 1 (2010).
[32] A. Alu and N. Engheta, Phys. Rev. B **75**, 024304 (2007).
[33] M. G. Silveirinha, Phys. Rev. B **76**, 245117 (2007).
[34] A. I. Kuznetsov, A. E. Miroshnichenko, Y. H. Fu, J. B. Zhang, and B. Luk'yanchuk, Sci. Rep. **2**, 492 (2012).
[35] J. A. Schuller, R. Zia, T. Taubner, and M. L. Brongersma, Phys. Rev. Lett. **99**, 107401 (2007).
[36] L. Shi, T. U. Tuzer, R. Fenollosa, and F. Meseguer, Adv. Mater. **24**, 5934 (2012).
[37] S. Zhang, R. Jiang, Y. M. Xie, Q. Ruan, B. Yang, J. Wang, and H. Q. Lin, Adv. Mater. **27**, 7432 (2015).
[38] B. I. Popa and S. A. Cummer, Phys. Rev. Lett. **100**, 207401 (2008).
[39] C. L. Holloway, E. F. Kuester, J. Baker-Jarvis, and P. Kabos, IEEE Trans. Antennas Propag. **51**, 2596 (2003).
[40] U. Zywietz, A. B. Evlyukhin, C. Reinhardt, and B. N. Chichkov, Nat. Commun. **5**, 3402 (2014).
[41] R. Collin, *Field Theory of Guided Waves* (IEEE, New York, 1991).
[42] E. Purcell and C. Pennypacker, Astrophys. J. **186**, 705 (1973).
[43] B. Draine, Astrophys. J. **333**, 848 (1988).
[44] A. V. Chebykin, M. A. Gorlach, and P. A. Belov, Phys. Rev. B **92**, 045127 (2015).
[45] W. J. Padilla, M. T. Aronsson, C. Highstrete, M. Lee, A. J. Taylor, and R. D. Averitt, Phys. Rev. B **75**, 041102 (2007).
[46] N. Kumar, A. C. Strikwerda, K. Fan, X. Zhang, R. D. Averitt, P. C. M. Planken, and A. J. L. Adam, Opt. Express **20**, 11277 (2012).
[47] C. Larsson, C. Sohl, M. Gustafsson, and G. Kristensson, in *Nordic Conference on Radio Science and Communications* (Vaxjo University Press, Vaxjo, 2008), p. 127.
[48] B. Gompf, J. Braun, T. Weiss, H. Giessen, M. Dressel, and U. Hubner, Phys. Rev. Lett. **106**, 185501 (2011).
[49] B. Gompf, B. Krausz, B. Frank, and M. Dressel, Phys. Rev. B **86**, 075462 (2012).