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Ultrafast non-thermal laser excitation of gigahertz longitudinal and shear acoustic waves in spin-crossover molecular crystals [Fe(PM-AzA)(2)(NCS)(2)]

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1 Ultrafast non-thermal laser excitation of gigahertz longitudinal and shear 2 acoustic waves in spin-crossover molecular crystals [$Fe(PM-AzA)_2(NCS)_2$]

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10 We report GHz longitudinal as well as shear acoustic phonon photoexcitation and photodetection
11 using femtosecond laser pulses in a spin-crossover molecular crystal. From our experimental obser-
12 vation of time domain Brillouin scattering triggered by the photoexcitation of acoustic waves
13 across the low-spin (LS) to high-spin (HS) thermal crossover, we reveal a link between the molecu-
14 lar spin state and photoexcitation of coherent GHz acoustic phonons. In particular, we experimen-
15 tally evidence a non-thermal pathway for the laser excitation of GHz phonons. We also provide
16 experimental insights into the optical and mechanical parameters evolving across the LS/HS spin
17 crossover temperature range. *Published by AIP Publishing.* <https://doi.org/10.1063/1.4996538>

18 Understanding how ultrafast photoinduced molecular
19 switching in crystals couples to the lattice in optical materi-
20 als is one of the key challenges in the fields of ultrafast
21 photo-induced phase transitions or transformations and ultra-
22 fast acoustics. Systems like spin-crossover compounds
23 exhibit, with temperature or pressure, change of the molecu-
24 lar spin state in the central d^4 - d^7 transition metals of the
25 complex. They are promising candidates for diverse applica-
26 tions including miniature temperature sensors, displays, data
27 storage, and photonic devices.^{1,2} Moreover, they can be fab-
28 ricated in a variety of forms (bulk, powders, matrices, and
29 films) and down sizable to nanoparticles.² The possibility to
30 trigger their properties by light,³ especially in an ultrafast
31 fashion, offers further prospects in future applications as
32 optically controlled switching devices.^{2,4} While such com-
33 pounds have already been widely studied,⁵⁻⁸ there is a short-
34 age of information about the interplay between the change of
35 the molecular spin state, the change of the unit cell volume
36 (5% change), and the covalent bonding (10% change). From
37 this standpoint, the study of crystal deformations that trigger
38 excitation of coherent acoustic phonons clearly deserves fur-
39 ther experimental investigations. Until now,⁹ in the field of
40 photo-induced phase transitions or transformations in molecu-
41 lar crystals, the role of coherent optical phonons has been
42 long under scrutiny because of the central role of optical
43 mode softening, whereas that of coherent acoustic phonons
44 and lattice deformations has not benefited from the same
45 surge of effort. It is of paramount importance from the
46 fundamental standpoint, as well as for the control of non-
47 volatile information and energy storage, to further explore
48 the pathways whereby acoustic phonons lead to non-volatile
49 photo-induced states.

50 In the following, we describe our experimental results
51 dealing with the photoexcitation of coherent acoustic phonons
52 in a spin-crossover crystal, performed at different temperatures

53 around the spin crossover temperature $T_{1/2}$. The possibility of
54 exciting with light pulse coherent acoustic phonons in a vari-
55 ety of materials is well known;^{10,11} however, unlike the longi-
56 tudinal acoustic phonons, the shear acoustic phonons are
57 difficult to photo-excite.^{12,13} Our results highlight peculiar and
58 efficient mechanisms for the photoexcitation of shear acoustic
59 phonons and shed light on the interplay between molecular
60 and elastic parameters in a spin-crossover compound.

61 In the present experiments sketched in Fig. 1(a),
62 spin-crossover molecular crystals [$Fe(PM-AzA)_2(NCS)_2$],
63 schematically represented in the inset of Fig. 1(b), were
64 investigated. These molecular crystals belong to the mono-
65 clinic space group $P2_1/c$ with one molecule as the asymmetric
66 unit. In agreement with Refs. 5 and 7, the smooth thermal
67 spin-crossover of these crystals with a spin crossover around
68 $T_{1/2} \sim 180$ K can be monitored experimentally from the mea-
69 surement of the magnetic susceptibility χ_M and the product
70 $\chi_M T$, as indicated in Fig. 1(b). The experiments were per-
71 formed on a parallelepipedic $4 \times 1 \times 1$ mm³ single crystal,
72 with smooth, black color surfaces.⁷ Due to large optical
73 absorption by the crystal, front side pump-probe transient
74 reflectivity measurements were performed, see Fig. 1(a). The
75 pump and probe beams originate from a femtosecond Ti-
76 Sapphire Coherent RegA 9000 regenerative amplifier operat-
77 ing at a central wavelength of 800 nm and delivering 160 fs
78 pulses at a repetition rate of 250 kHz. The 400 nm pump
79 pulses of about 40 nJ energy per pulse were focused on the
80 (110) surface of the crystal with a gaussian spatial profile of
81 FWHM ~ 100 μ m. The time delayed 800 nm probe of tenfold
82 weaker energy per pulse, vertically polarized, was tightly
83 focused at 30° oblique incidence on the (110) surface normal
84 to the crystal and spatially overlapped with the pump spot.
85 The reflected probe beam was directed to a photodiode cou-
86 pled to a lock-in amplifier operating at 50 kHz of the pump
87 laser modulation frequency, to measure transient differential
88 reflectivity $\Delta R(t)$ as a function of time delay between pump
89 and probe beams. Upon transient absorption of the 400 nm
90 pump pulse over the optical skin depth of the crystal, the light

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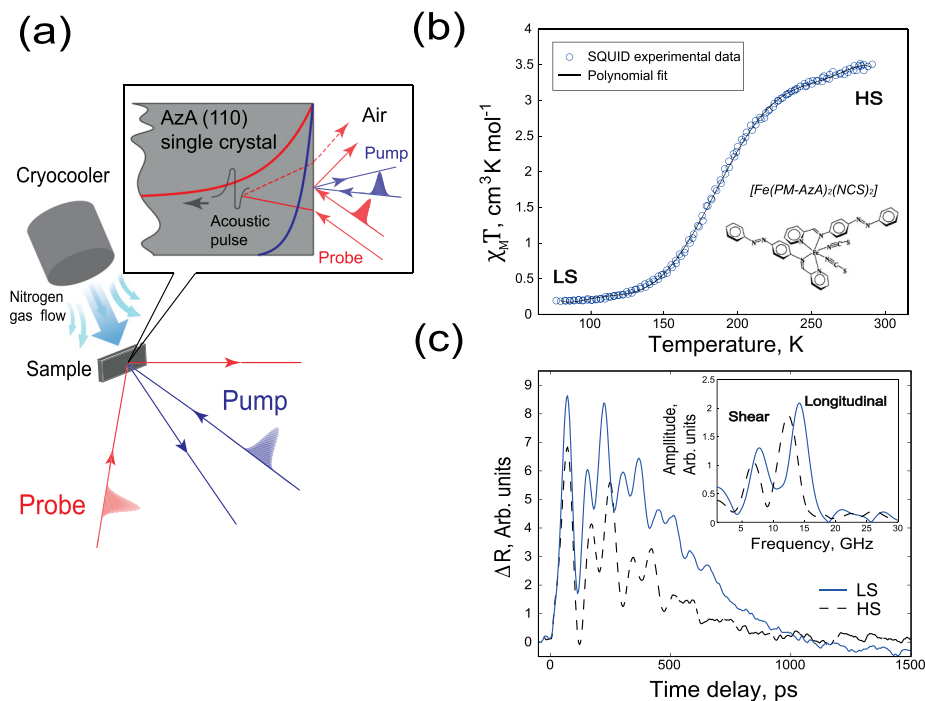


FIG. 1. (a) Sketch of the experimental setup. A laser pump pulse photoexcites the molecular crystal sample under continuous nitrogen flow for temperature control. The propagation of the photoexcited acoustic pulse is detected by a time-delayed optical probe pulse vertically polarized. (b) Schematic representation of the spin-crossover compound⁵ and its magnetic susceptibility recorded across the smooth thermal spin-crossover temperature $T_{1/2}$. (c) Transient reflectivity signals recorded in the LS and HS states. The frequency spectrum of the LS and HS Brillouin oscillations shown in the inset gathers crucial information on the photoexcitation of both longitudinal and shear acoustic phonons.

91 energy is partially converted into mechanical energy that
 92 drives the excitation of acoustic pulses propagating away
 93 from the free surface. In the present situation, the crystal is
 94 about five times less absorptive at 800 nm than at 400 nm,
 95 see Fig. S1 in the [supplementary material](#), such that it can
 96 be considered as semi-transparent at 800 nm and opaque at
 97 400 nm. As a consequence, the pump light is locally absorbed
 98 at the free surface where it launches a propagating acoustic
 99 strain that backscatters the probe light from within the semi-
 100 transparent medium and leads to the occurrence of time
 101 domain Brillouin scattering oscillations, see Fig. 1(c). As in
 102 any Brillouin scattering process, the frequency ν of these
 103 oscillations is related to the ultrasound velocity v of the crystal,
 104 to the probe wavelength λ , to the refractive index n of the
 105 medium, and to the back-scattering angle θ through

$$\nu = 2 n v \cos \theta / \lambda. \quad (1)$$

106 The light activation of both longitudinal and shear acoustic
 107 polarizations, of different ultrasonic speeds v , leads to two
 108 distinct Brillouin frequencies. Figure 1(c) shows an example
 109 of such time domain Brillouin scattering light modulation
 110 where unambiguous periodic features at about 6 GHz and
 111 12 GHz, see inset, are evidenced right after pump excitation
 112 at zero time delay. The shear acoustic nature of the 6 GHz
 113 frequency has been further experimentally confirmed from
 114 depolarized Brillouin scattering measurements, see Fig. S2
 115 in the [supplementary material](#), which enhances the optical
 116 detection of shear acoustic waves.¹³

117 The experiments have been conducted at different tem-
 118 peratures ranging from 100 K, where almost 100% of mole-
 119 cules are in the low spin state, up to 300 K, where almost
 120 100% of the molecules are in the high-spin state. Temperature
 121 steps of 10 K were performed with a continuous nitrogen
 122 flow. For each recorded time domain Brillouin scattering sig-
 123 nal in the 100–300 K temperature range, we have numerically
 124 fitted the experimental data with a damped sinusoidal function

in the form $\sim A \exp(-t/\tau) \sin(2\pi\nu t + \phi)$, to retrieve the fre- 125
 quency ν , the damping time τ , the amplitude A , and the phase 126
 ϕ of each longitudinal and shear acoustic mode. Based on the 127
 results from ellipsometry available in the [supplementary mate- 128](#)
 rial, which led to the observation of a slight variation of only a 129
 few percent of the optical refractive index of the crystal at dif- 130
 ferent temperatures, the huge 15% change in Brillouin fre- 131
 quency in Fig. 2(a) is mainly due to a pronounced change in 132
 both longitudinal and shear acoustic speeds with temperature. 133
 According to Eq. (1) and the measured real part n_{\perp} of the 134
 index of refraction at the probe wavelength for vertically polar- 135
 ized light, see [supplementary material](#), we have computed the 136
 change in Brillouin frequency to extract the variation in longi- 137
 tudinal and shear acoustic speed with temperature. The results 138
 displayed in Fig. 2(b) confirm the substantial change in acous- 139
 tic speed, in the range of 10%, further emphasizing the giant 140
 change in mechanical properties of the material across the spin 141
 crossover temperature. This intrinsic softening of the material 142
 at increasing temperature around $T_{1/2}$ is coupled to a substan- 143
 tial isostructural modification of the lattice parameters, mani- 144
 fested in the change of the unit cell volume by as much as 145
 $\sim 3\%$.^{5,14} 146

As a comparison with highly magnetostrictive ferromag- 147
 netic compounds such as Terfenol¹⁵ which is the foremost 148
 highest magnetostrictive alloy with a change of the unit cell 149
 volume by an amount of 0.1% upon modification of the mag- 150
 netization vector, the change in $[Fe(PM-AzA)_2(NCS)_2]$ lat- 151
 tice parameters in the order of few % reveals the gigantic 152
 spin state-lattice coupling in these molecular materials. The 153
 pronounced molecular spin state-lattice coupling in such 154
 compounds is bound to influence the generation of coherent 155
 acoustic phonons. 156

The evolutions of the damping time coefficients with 157
 temperature displayed in Fig. 2(c) are linked to the imagi- 158
 nary part of the refractive index k_{\perp} at the probe wavelength 159
 and to the intrinsic acoustic attenuation. From the measured 160
 k_{\perp} at the probe wavelength, we have processed the measured 161

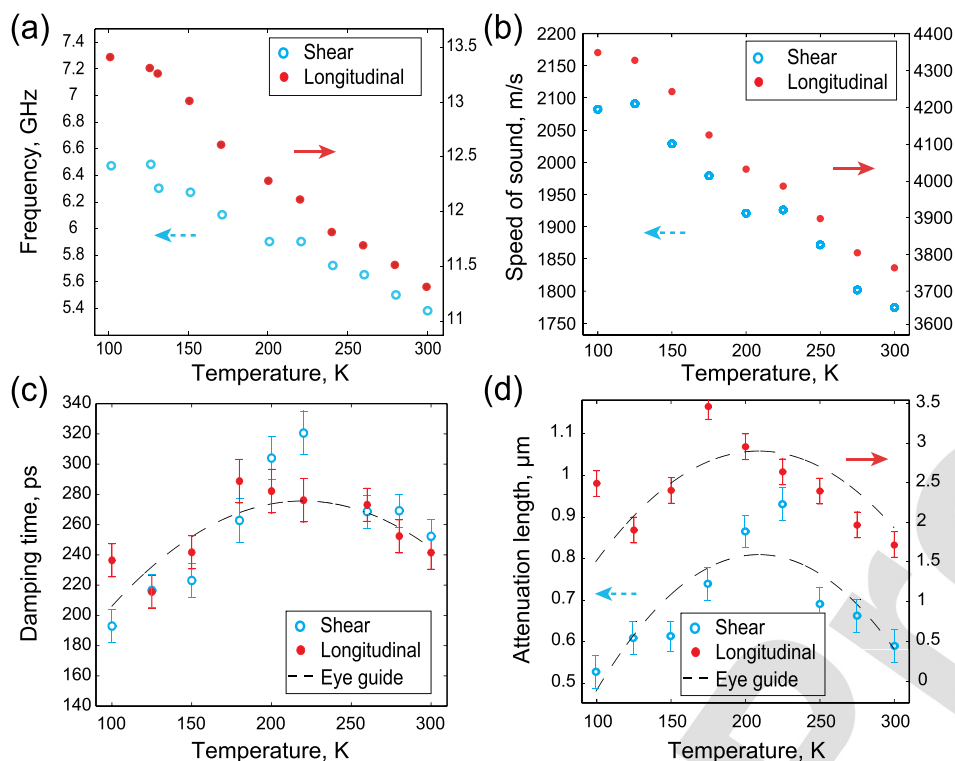


FIG. 2. From the time domain Brillouin scattering experimental data obtained for vertically polarized pump and probe beams at different temperatures, we have extracted the frequency (a) and the damping coefficient (c) of the measured Brillouin oscillations. In a second step, the temperature evolution of the index of refraction in the [supplementary material](#) led to the calculated acoustic speed (b) and acoustic attenuation length (d) of the longitudinal and shear acoustic modes across $T_{1/2}$. The eye guides are 2nd order polynomial fit of the extracted coefficients and the error bars are estimated from our experimental uncertainties.

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162 damping of the Brillouin oscillations τ , in order to extract
 163 the acoustic attenuation length Γ straightforwardly from
 164 $\Gamma = v\tau - \zeta$, where v is the longitudinal or shear acoustic speed
 165 and $\zeta = 4\pi k_{\perp}/\lambda$ is the optical penetration depth. Note that
 166 the deconvolution of the acoustic damping in the measured
 167 signals is not as straightforward in classical Brillouin spectroscopy
 168 which sometimes requires complicated analyses
 169 of the Brillouin linewidth of the peaks in the frequency
 170 domain.¹⁶ The result of the calculation of the acoustic attenuation
 171 length for both acoustic modes, longitudinal and shear,
 172 is displayed in Fig. 2(d). The remarkable maximum attenuation
 173 length measured in Fig. 2(d) highlights a decrease in the
 174 acoustic attenuation across the spin-phase crossover transition.
 175 In the present case, the phenomenon is reversed from
 176 the well-known structural α -relaxation which has been evidenced
 177 in glass forming liquids across the T_g glass transition
 178 temperature.¹⁷ The acoustic wave propagates on longer distances
 179 at the spin-crossover temperature which indicates that
 180 the structural modification of the lattice and the statistical
 181 growth or disappearance of the LS/HS states do not perturb
 182 the acoustic phonon propagation; on the contrary, the acoustic
 183 phonon propagation is facilitated during the spin crossover
 184 transition.

185 We can invoke several main mechanisms for the laser-driven
 186 lattice motion in the present spin-crossover material:
 187 the thermoelastic mechanism which is linked to the transient
 188 thermal dilation of the lattice following the temperature rise
 189 due to laser absorption and two non-thermal mechanisms,
 190 namely, the molecular spin state-lattice coupling mechanism¹⁸
 191 and the deformation potential mechanism.¹³ The temperature
 192 evolution of the Brillouin amplitudes and Brillouin phases
 193 at two different pump polarizations [horizontal (H) and vertical (V)
 194 polarizations] displayed in Fig. 3 gathers crucial information
 195 on the photoacoustic excitation process.
 196 The fact that the optical index of refraction does not vary

197 significantly with temperature warrants that the measured
 198 Brillouin amplitude and phase are mainly sensitive to the
 199 excitation mechanisms and not to the detection process
 200 through a modification of the acousto-optic coefficients with
 201 temperature. Since the measurement of the Brillouin amplitude
 202 can suffer from experimental artifacts, such as beam
 203 pointing stability during sample heating or cooling, we have
 204 chosen to further process the longitudinal A_L and shear A_S
 205 Brillouin amplitudes data by taking the ratio of both amplitudes,
 206 defined as A_L/A_S , not biased by optical artifacts. This
 207 simple procedure highlights a discrepancy shown in Fig. 3(a)
 208 that cannot be assigned to the thermoelastic mechanism.
 209 In fact, as indicated by the difference in optical absorption
 210 coefficients k_{\perp} and k_{\parallel} at the pump wavelength, see [supplementary material](#),
 211 the laser induced temperature rise in such anisotropic crystals
 212 depends on the pump polarization.
 213 However, taking the ratio of amplitudes of both modes is a
 214 proper way to conveniently remove the temperature rise
 215 contribution in the thermoelastic process of acoustic excitation,
 216 similar for longitudinal and shear acoustic modes. Therefore,
 217 the gap between the amplitude ratio in Fig. 3(a) with H or V
 218 polarizations is assigned to a non-thermal mechanism, either
 219 molecular spin state-lattice coupling or deformation potential
 220 mechanism. One possible explanation of the observed amplitude
 221 jump would be the anisotropy in the deformation potential
 222 mechanism. The latter should be considered as tensorial
 223 in such crystals with different diagonal and non-diagonal
 224 coefficients referring to preferential electronic excitations
 225 of the ligands by horizontally or vertically polarized pump
 226 pulses, with some similarities with Ref. 19 that demonstrates
 227 the wide range of electronic excitations of organic molecules
 228 that can drive coherent lattice phonon excitation. In addition
 229 to the electron deformation potential mechanism, we cannot
 230 neglect the molecular spin state-lattice coupling anisotropy,
 231 thus revealing pump laser polarization dependence.

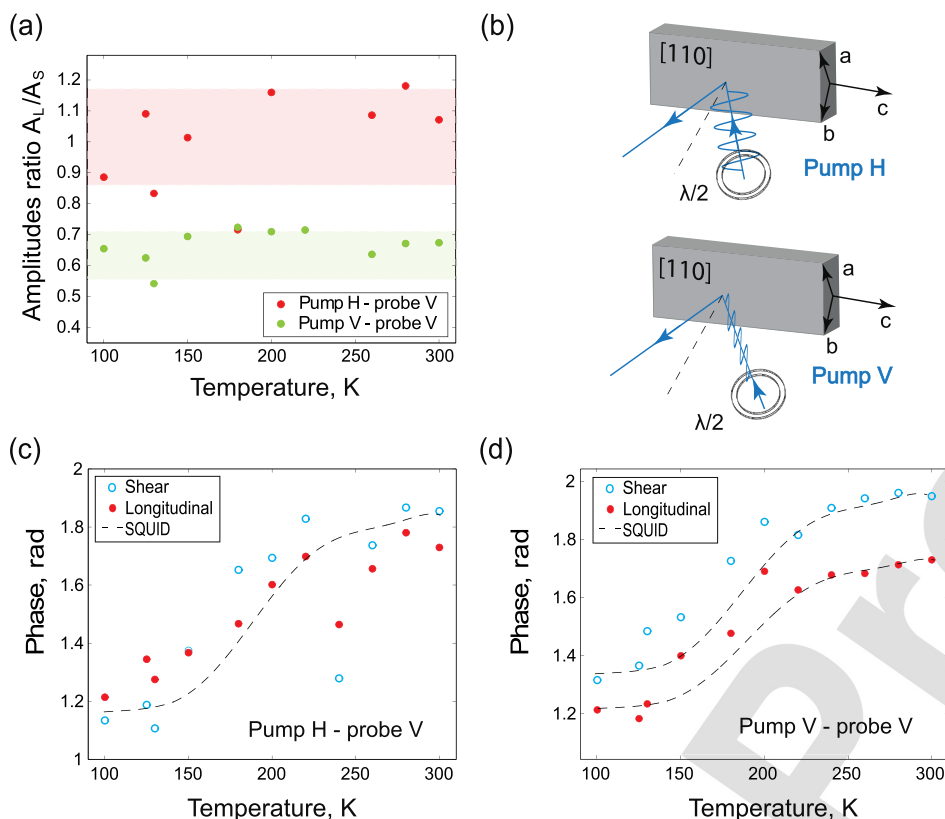


FIG. 3. (a) Ratio between the longitudinal and shear Brillouin amplitudes for different pump polarizations. Red dots correspond to the horizontally polarized pump and green to the vertically polarized pump, as sketched in (b). Extracted time domain Brillouin phase for horizontally (c) and vertically (d) polarized pump beams of both longitudinal and shear acoustic modes in the compound at different crystal temperatures. The SQUID curve matches the Brillouin phase evolution across $T_{1/2}$.

Another striking feature suggesting coexistence of photoacoustic mechanisms is revealed by the Brillouin phase change in Figs. 3(c) and 3(d) with H or V pump polarizations. Once again, if we assume that the slight change with temperature of the optical index of refraction is irrelevant for the interpretation of our experimental observations, the substantial phase jump in the range of 0.6 radians of both longitudinal and shear Brillouin signals across the spin-crossover transition highlights a profound change in the laser-matter mechanism for acoustic phonons excitation. Based on Ref. 20, we can interpret this phase change, which correlates with the magnetic susceptibility of Fig. 1(b), as a change in the acoustic excitation process through the contribution of the spin state-lattice mechanism that vanishes once the compound reaches 100% HS spin. However, the photoinduced spin state-lattice coupling is maybe not the most efficient at this pump wavelength. Therefore, we cannot rule out the deformation potential mechanism as relevant in the process of laser excitation of coherent acoustic phonons in the present spin crossover material. The observation of the phase jump of about 0.2 radians in Fig. 3(d) between longitudinal and shear excitation points to a non-thermal mechanism of acoustic excitation sensitive to the pump polarization, as in Fig. 3(a). As a matter of fact, based on the results presented in Fig. 3, we can conclude that two non-thermal mechanisms are triggered in these molecular crystals, each one having different efficiency (amplitudes) and characteristic times (phases).

In summary, we have performed ultrafast time-domain Brillouin scattering experiments to study non equilibrium dynamics following femtosecond photoexcitation of spin-crossover molecular crystals $[Fe(PM-AzA)_2(NCS)_2]$. We have presented results for coherent GHz acoustic phonons photo-generation and photodetection in a spin-crossover material

across the spin-crossover temperature range. Through time domain Brillouin scattering, we evidence non-thermal excitation of acoustic phonons which are of spin state-lattice coupling and/or deformation potential origin. Experimentally revealed on the sub-nanosecond time-scale, remarkable sensitivity of Brillouin frequencies to the spin state of a molecular material opens advanced perspectives for probing macroscopically relevant processes during a phase transition. We envisage pump-pump-probe experiments, in which the molecular spin-state is photoexcited with wavelength tuned pump pulse, while the real-time coupling of thus generated spin-states to the lattice is followed by the second pump through time-resolved Brillouin scattering, such as this work. Furthermore, our results highlight the versatile and efficient generation of ultrashort shear acoustic phonons for future investigations of viscoelastic properties of materials such as liquids,²¹ glasses,^{22,23} mixed multiferroics, correlated electron systems, and magnetic materials. Ultimately, deeper knowledge of the spin state-elastic coupling in spin-crossover molecular crystals will be crucial for the design of multifunctional molecular devices.

See supplementary material for the temperature data of the real n and imaginary k refractive index of $[Fe(PM-AzA)_2(NCS)_2]$ at 400 nm and 800 nm wavelengths, as for a comparison between transient reflectivity and depolarized Brillouin scattering.

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