LA Phonoritons in Cu_2O

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We report the observation of LA-phonoriton eigenstates, which are a pump-induced coherent superposition of excitons, photons, and longitudinal acoustic (LA) phonons developing in up-conversion Brillouin scattering of an intense polariton. The excitonic component of the LA phonoritons in a Cu_2O bulk crystal in the presence of a coherent orthoexcitonic polariton is studied by stimulated two-photon emission spectroscopy. For optical intensities of $30-200$ MW/cm^2 the relevant scale of the pump-induced spectral changes is $5-10$ μeV. The theory of LA phonoritons in Cu_2O quantitatively reproduces the experimental data.

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Since the discovery of an exciton in Cu_2O [1] this semiconductor became a prototype material in the physics of excitons, due to the large binding energy of the ground-state exciton and due to an extremely high quality of the naturally grown crystals. A ground-state $1S$ orthoexciton of the yellow series (binding energy $\epsilon^x \simeq 150$ meV and Bohr radius $a_x \simeq 7$ Å) is dipole forbidden. However, at low temperatures the quadrupole interaction between the orthoexciton and the light field leads to a weak, but still well-defined, polariton [2]. Recently, the quadrupole polaritons in Cu_2O were finally visualized by propagation beat spectroscopy [3].

The polariton is an example of a linear mode (coherent superposition of exciton and photon). Usually, this contrasts with purely scattered, incoherent states in, e.g., Raman and Brillouin scattering of polaritons [4–6]. However, when pumped by an intense laser, up-conversion phonon-assisted scattering can reveal a *phonoriton* spectrum [7–11]. It is a quasilinear mode depending parametrically on the intensity of the pump light. It characterizes a coherent superposition of exciton, photon, and phonon. The phonoriton spectrum associated with optical phonons gives rise to the phonon-mediated optical Stark effect, which has already been observed in some polymers (e.g., polydiacetylene) as well as in the inorganic semiconductors CdS and HgI_2 [12]. In this Letter we report the first observation of longitudinal acoustic (LA) phonoritons. For this purpose we study stimulated two-photon emission (STPE) from *virtual excitons* created by up-conversion Brillouin scattering of a high-intensity quadrupole polariton in Cu_2O. Because of the linear dispersion of acoustic phonons the resulting dispersions are different from those of optical phonoritons. Furthermore, the thermally activated LA phonoritons contribute to STPE even at helium temperatures.

The quadrupole polaritons in Cu_2O are characterized by the dispersion equation $(\omega_{\mathbf{p}}^{\gamma}/\omega)^2 - 1 = \Omega_c^2(p)/[(\omega_{\mathbf{p}}^{x})^2 - \omega^2]$, where $\omega_{\mathbf{p}}^{x} = \omega_t + \hbar p^2/2M_x$ and $\omega_{\mathbf{p}}^{\gamma} = cp/\sqrt{\varepsilon_b}$ are the orthoexciton and photon dispersions, respectively, M_x is the exciton translational mass, $\hbar\omega_t$ is the energy of an orthoexciton at rest, $\hbar\mathbf{p}$ is the exciton momentum, and ε_b is the background dielectric constant. The polariton parameter is $\Omega_c(p) = f^{1/2}\omega_p^{\gamma} \propto p$, where $f \simeq 3.7 \times 10^{-9}$ is the dimensionless oscillator strength of the orthoexciton-photon interaction [3]. The spectral separation between the upper $\omega_{\mathbf{p}}^{+}$ and lower $\omega_{\mathbf{p}}^{-}$ polariton dispersion branches at the resonant wave vector $k_R \simeq 2.62 \times 10^5$ cm^{-1} is given by $\omega_{k_R}^{+} - \omega_{k_R}^{-} = \Omega_c(k_R) \simeq 124$ μeV. The dephasing rate Γ^x of orthoexcitons in Cu_2O is very low, i.e., $\Gamma^x \leq 1$ μeV at temperatures $T \leq 2$ K [3,6]. Because of the weak exciton-exciton interaction, estimated by $U_0 = 4\pi\hbar^2 a_x/M_x \simeq 2.8 \times 10^{-22}$ eV cm^3, up to a high concentration of orthoexcitons $N_0^x \sim 10^{18}$ cm^{-3} the main scattering mechanism is the exciton-LA-phonon deformation potential interaction [13]. Because $2\hbar v_s k_R \simeq 155$ μeV is comparable to $\Omega_c(k_R)$ (v_s is the LA-sound velocity), the quadrupole polariton with $\omega_{\mathbf{p}}^{-} < \omega_t$ undergoes mainly anti-Stokes Brillouin scattering.

An intense laser pulse with the frequency $\omega = \omega_{\mathbf{k}}$ below $\omega_t - \mathrm{Max}\{\Gamma^x, \Omega_c^2(k_R)/2\omega_t\}$ induces a *macroscopic or pump polariton* with the exciton wave vector $\mathbf{k}$ given by the equation $\omega_{\mathbf{k}} = \omega_{\mathbf{k}}^{-}$. The excitonic component of the macroscopic polariton keeps the coherence of the incoming pump light and couples with other excitonic (polariton) modes $\mathbf{p}$, due to the exciton-LA-phonon interaction. The concentration of pump-induced virtual orthoexcitons $\mathbf{k}$ is given by $N_0^x = (I_0/\hbar\omega_{\mathbf{k}})(\sqrt{\varepsilon_b}/c)[\Omega_c^2(k)/4(\omega_t - \omega_{\mathbf{k}})^2]$, where I_0 is the intensity of the pump light. In the presence of a quasi-cw macroscopic quadrupole polariton $(\mathbf{k}, \omega_{\mathbf{k}})$, the orthoexciton-photon-phonon Hamiltonian of a Cu_2O crystal is approximated by

$$H = \sum_{\mathbf{p}} \hbar\Big[(\omega_{\mathbf{p}}^{x} - \omega_{\mathbf{k}}) B_{\mathbf{p}}^{\dagger} B_{\mathbf{p}} + (\omega_{\mathbf{p}}^{\gamma} - \omega_{\mathbf{k}}) \alpha_{\mathbf{p}}^{\dagger} \alpha_{\mathbf{p}} + \Omega_{\mathbf{p}-\mathbf{k}}^{\mathrm{ph}} c_{\mathbf{p}-\mathbf{k}}^{\dagger} c_{\mathbf{p}-\mathbf{k}} + i\frac{\Omega_c(p)}{2}(\alpha_{\mathbf{p}}^{\dagger} B_{\mathbf{p}} - B_{\mathbf{p}}^{\dagger} \alpha_{\mathbf{p}}) + iQ(\mathbf{p} - \mathbf{k})(B_{\mathbf{p}}^{\dagger} c_{\mathbf{p}-\mathbf{k}} - c_{\mathbf{p}-\mathbf{k}}^{\dagger} B_{\mathbf{p}}) \Big], \quad (1)$$

where $B_{\mathbf{p}}$, $\alpha_{\mathbf{p}}$, and $c_{\mathbf{p}-\mathbf{k}}$ are the exciton, photon, and LA-phonon operators, respectively, and $\Omega^{\rm ph}_{\mathbf{p}-\mathbf{k}} = v_s|\mathbf{p} - \mathbf{k}|$ is the LA-phonon dispersion. The matrix element Q of the pump-induced orthoexciton-LA-phonon interaction is given by $Q(\mathbf{p} - \mathbf{k}) = D_x[|\mathbf{p} - \mathbf{k}|/(2\hbar\rho v_s)]^{1/2}\sqrt{N_0^x}$, where D_x is the exciton deformation potential, and ρ is the mass density. The approximation of the exciton-phonon interaction by the last term on the right-hand side of Eq. (1) means that we include only the orthoexciton-LA-phonon coupling mediated by the pump polariton.

The quadratic Hermitian Hamiltonian (1) describes the LA-phonoriton eigenstates of bulk Cu_2O virtually excited by coherent light with the frequency $\omega_{\mathbf{k}} \leq \omega_t$. From Eq. (1), the phonoriton dispersion equation is

$$(\omega^x_{\mathbf{p}} - \omega)(\omega^\gamma_{\mathbf{p}} - \omega)(\omega_{\mathbf{k}} + \Omega^{\rm ph}_{\mathbf{p}-\mathbf{k}} - \omega) - Q^2(\mathbf{p} - \mathbf{k})(\omega^\gamma_{\mathbf{p}} - \omega) - \frac{\Omega_c^2(p)}{4}(\omega_{\mathbf{k}} + \Omega^{\rm ph}_{\mathbf{p}-\mathbf{k}} - \omega) = 0. \quad (2)$$

Two basic parameters $\Omega_c(p)$ and $Q(\mathbf{p} - \mathbf{k}) \propto \sqrt{I_0}$, which characterize the exciton-photon quadrupole interaction and the pump-induced anti-Stokes transitions, respectively, enter Eq. (2). With decreasing pump intensity I_0, the phonon coupling strength $Q(\mathbf{p} - \mathbf{k})$ vanishes and the LA phonoriton $\mathbf{p}$ decouples into the polariton $\mathbf{p}$ and LA phonon $\mathbf{p} - \mathbf{k}$. The LA-phonoriton spectrum consists of three dispersion branches $\omega = \omega_{i=1,2,3}(\mathbf{p}, I_0)$ (see Fig. 1; $i = 1, 2, 3$ refer to the upper, middle, and lower pump-induced LA-phonoriton dispersion branches, respectively) and originates from the mutual hybridization of the initial orthoexciton, photon, and LA-phonon dispersions, similarly to how the polariton dispersion develops from the exciton and photon spectra.

The LA-phonoriton spectrum can be interpreted in the following way. The pump quadrupole polariton $\mathbf{k}$ absorbs a thermal LA phonon with momentum $\mathbf{p} - \mathbf{k}$. As a result, a virtual orthoexciton with wave vector $\mathbf{p}$ is created by anti-Stokes scattering. In turn, the created exciton can undergo a stimulated Stokes transition back into the mode $\mathbf{k}$, assisted by the emission of the initial LA phonon $\mathbf{p} - \mathbf{k}$. The coherent LA-phonon-mediated oscillations between the scattered modes $\mathbf{p}$ and the pump-driven mode $\mathbf{k}$ result in the development of a split at the anti-Stokes resonance of the pump polariton (see Fig. 1). In the general case, the phonoriton eigenstate $\mathbf{p}$ originates from the complicated pump-induced coherent nutations between the three states, exciton $\mathbf{p}$, LA phonon $\mathbf{p} - \mathbf{k}$, and photon $\mathbf{p}$. The standard picture of phonon-mediated polariton scattering implies that the up-converted Brillouin replicas of the scattered polariton $\mathbf{k}$ arise at the spectral positions given by the equation $\omega^{\pm}_{\mathbf{p}} = \omega_{\mathbf{k}} + \Omega^{\rm ph}_{\mathbf{p}-\mathbf{k}}$, due to energy-momentum conservation. In contrast, the phonoriton eigenstate allows a pump-induced virtual excitonic component at spectral points $[\mathbf{p}, \omega_i(\mathbf{p})]$ outside the polariton dispersion.

The STPE method detects the excitonic occupation of the mode $\mathbf{p} = \mathbf{k}_{\rm pr} + \mathbf{k}_{\rm sig}$ [14]. The STPE signal at wave vector $\mathbf{k}_{\rm sig}$ and frequency $\omega_{\rm sig}$ is induced by the probe light $(\mathbf{k}_{\rm pr}, \omega_{\rm pr})$. The orthoexciton state Γ_5^+ is threefold degenerate. According to the quadrupole selection rules, only the $\Gamma_{5-}^+ \equiv (1/\sqrt{2})(\Gamma_{5x}^+ - \Gamma_{5y}^+)$ component of the orthoexciton is excited for $\mathbf{k}\|(111)$ and polarization along $(1\bar{1}0)$. The probe light with $\mathbf{k}_{\rm pr}\|(111)$ and polarization along $(11\bar{2})$ induces the STPE signal with $\mathbf{k}_{\rm sig}\|(111)$ and polarization along $(1\bar{1}0)$ in the forward-scattering direction (see the inset of Fig. 2). The above geometry corresponds to our model, which operates with the one-component orthoexciton.

Intense light pulses of 7 ns duration at $\lambda = 1064$ nm were generated by a Nd:YAG laser. A dye laser with

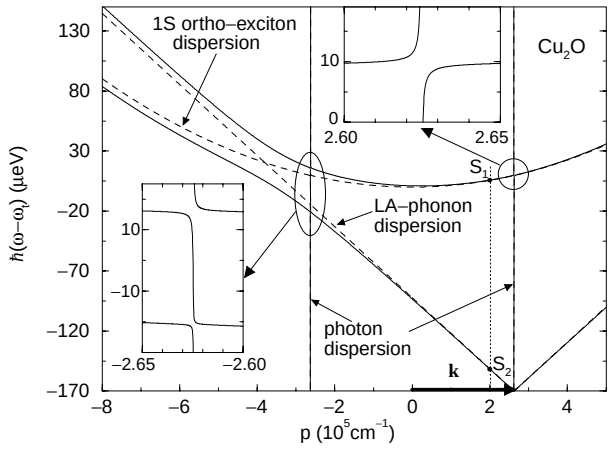

FIG. 1. Dispersion of LA phonoritons in Cu_2O (solid lines, $N_0^x = 10^{18}$ cm^{-3}). The bold arrow indicates the pump polariton $[\hbar(\omega_{\mathbf{k}} - \omega_t) = -170\ \mu$eV]. Dashed lines, orthoexciton and LA-phonon dispersions.

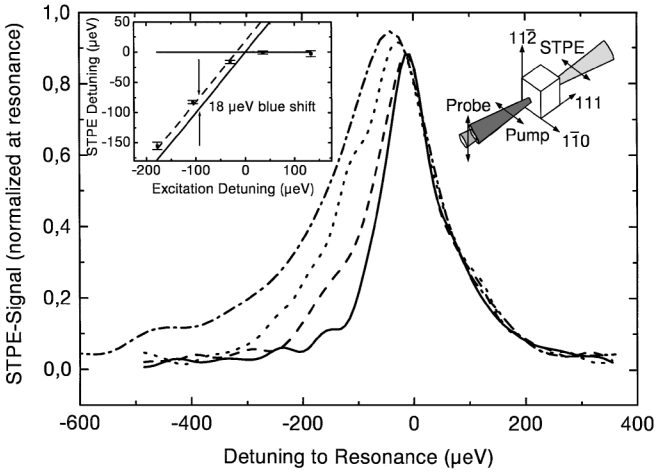

FIG. 2. The STPE signal for the intensity of the pump light $I_0 = 30$ MW/cm^2 (solid line), 45 MW/cm^2 (dashed line), 60 MW/cm^2 (dotted line), and 200 MW/cm^2 (dot-dashed line); $T = 1.9$ K. The spectra are normalized at the spectral position of the maximum signal at low I_0 (zero detuning, i.e., $\delta = \omega^x_{\mathbf{p}_0} - \omega_{\mathbf{k}} - \Omega^{\rm ph}_{\mathbf{p}_0-\mathbf{k}} = 0$). Left inset: $\omega_t - \omega_{\rm sig} - \omega_{\rm pr}$ vs $\omega_t - \omega_{\mathbf{k}}$. Right inset: the STPE geometry.

Sulforhodamine B was pumped by the second harmonic and served as pump laser which can scan the $1S$ orthoexciton at $\hbar\omega_t = 2.0327$ eV. The probe beam at $\hbar\omega_{\rm pr} = 0.6496$ eV was gained by Raman shifting the YAG laser in H_2 gas. Both the dye laser and Raman laser were focused on the sample to produce the STPE signal in the forward-scattering geometry. Using a double 0.85 m monochromator, the STPE signal was selected and recorded by a N_2-cooled CCD camera [$\hbar\omega_{\rm sig} \simeq \hbar(\omega_t - \omega_{\rm pr}) \simeq 1.383$ eV] with spectral resolution of about 6 μeV. The frequency-integrated STPE signal was detected by a GaAs photomultiplier. We used a naturally grown Cu_2O crystal with a quadrupole absorption line of 80 μeV width at $T = 1.9$ K.

The STPE signal from the Cu_2O crystal in the presence of the macroscopic polariton $(\mathbf{k}, \omega_{\mathbf{k}})$ induced by the pump light of various I_0 is shown in Fig. 2. According to the inset of Fig. 2, for negative detunings ($\omega_{\mathbf{k}} - \omega_t < 0$), one has $\hbar(\omega_{\rm pr} + \omega_{\rm sig} - \omega_{\mathbf{k}}) \simeq 18$ μeV. From this we specify the STPE-probed mode $\mathbf{p} = \mathbf{p}_0$ along (111) by $p_0 \simeq 2.02 \times 10^5$ cm^{-1}. Basically, three spectral points $S_{i=1,2,3} = [\mathbf{p}_0, \omega_{i=1,2,3}(\mathbf{p}_0)]$ of the LA-phonoriton dispersion can contribute to the STPE signal. However, the spectral point S_3 has no excitonic component, because $\hbar[\omega_t - \omega_{i=3}(\mathbf{p}_0)] \simeq 0.47$ eV $\gg \Omega_c(k_R)$. Thus only the upper and middle phonoriton dispersion branches can potentially contribute to the STPE signal (see the spectral points S_1 and S_2 in Fig. 1).

The photon component of the phonoriton state $\mathbf{p}_0$ is approximated by $\phi^{\gamma}_{i=1} = [\Omega_c(p_0)/4\Delta]^2$ and $\phi^{\gamma}_{i=2} = (Q_0/\delta)^2\phi^{\gamma}_{i=1}$, where $\Delta = \omega^x_{\mathbf{p}_0} - \omega^{\gamma}_{\mathbf{p}_0}$, $Q_0 = Q(\mathbf{p}_0 - \mathbf{k})$, and $\delta = \omega^x_{\mathbf{p}_0} - \omega_{\mathbf{k}} - v_s|\mathbf{p}_0 - \mathbf{k}|$ ($\hbar\delta$ is the energy separation between the spectral points S_1 and S_2, see Fig. 1). Because $\phi^{\gamma}_{i=2}(\mathbf{p}_0) \ll \phi^{\gamma}_{i=1}(\mathbf{p}_0) \simeq 1.6 \times 10^{-8}$, the photon component is negligible and the LA-phonoriton dispersion Eq. (2) reduces to

$$(\omega_{\mathbf{k}} + \Omega^{\rm ph}_{\mathbf{p}_0-\mathbf{k}} - i\Gamma^{\rm LA} - \omega)(\omega^x_{\mathbf{p}_0} - i\Gamma^x - \omega) = Q_0^2, \tag{3}$$

where the dephasing rates of LA phonons and orthoexcitons, $\Gamma^{\rm LA}$ and Γ^x, are included. The quadratic Eq. (3) describes the phonoritons of the dispersion branches $i = 1, 2$ as a quantum superposition of the exciton and phonon states. The interaction strength Q_0^2 between the excitonic and LA-phonon dispersions is proportional to $D_x^2\Omega_c^2(k)I_0$, i.e., the quadrupole polariton effect refers only to the pump light dressed by virtual excitons $\mathbf{k}$.

The lifetime of a long wavelength LA phonon with wave vector $\mathbf{q}$ originates from the lattice anharmonicity and can be estimated by $\tau^{\rm LA} = 1/\Gamma^{\rm LA} \simeq (\hbar^3\rho v_s^4)/[(k_BT)^4 q]$ [15]. The estimate yields $\tau^{\rm LA}(\mathbf{p}_0 - \mathbf{k})$ on a ms time scale, i.e., $\Gamma^{\rm LA}/\Gamma^x \sim 10^{-6}$. For the spectrally well-separated terms $\omega^x_{\mathbf{p}_0}$ and $\omega_{\mathbf{k}} + \Omega^{\rm ph}_{\mathbf{p}_0-\mathbf{k}}$, when $\delta = \overline{S_1S_2} \gg Q_0, \Gamma^x$, the phonoriton frequencies are given by $\omega_{i=1}(\mathbf{p}_0) = \omega^x_{\mathbf{p}_0} + Q_0^2/\delta - i\Gamma^x$ and $\omega_{i=2}(\mathbf{p}_0) = \omega_{\mathbf{k}} + \Omega^{\rm ph}_{\mathbf{p}_0-\mathbf{k}} - Q_0^2/\delta - i\Gamma^{\rm LA} - i\Gamma^x(Q_0/\delta)^2$. For intensities in the range 10 MW/cm$^2 \le I_0 \le 200$ MW/cm^2, the pump-induced repulsion $2Q_0^2/\delta$ between the two frequencies is much less than Γ^x, but much larger than $\Gamma^{\rm LA} + \Gamma^x(Q_0/\delta)^2$. This means that for the STPE-probed upper polariton dispersion branch (see point S_1 in Fig. 1) the phonoriton effect is relaxed and removed by dephasing, while for the spectral vicinity of S_2 the phonoriton eigenstate is well defined. For the resonant condition $\omega_{\mathbf{k}} + \Omega^{\rm ph}_{\mathbf{p}_0-\mathbf{k}} = \omega^x_{\mathbf{p}_0}$, when the spectral points S_1 and S_2 coincide ($\delta = 0$), the STPE signal relates to the pump-induced phonoriton split.

The STPE method is very suitable to study phonoritons, because it tests the I_0-dependent excitonic component of the phonoriton $\mathbf{p}_0$ rather than the pump-induced spectral changes, which occur on a few μeV energy scale. The inset of Fig. 2 shows that, for negative detunings of the pump light from the excitonic resonance, the STPE signal refers to the phonoriton dispersion branch $i = 2$, i.e., to the spectral point S_2. In contrast, for positive detunings of the pump light ($\omega_{\mathbf{k}} - \omega_t > 0$) the STPE signal stems from the upper dispersion branch, i.e., from the spectral point S_1 (see the inset of Fig. 2). This is because for positive detunings the efficiency of Stokes scattering is very high. The latter is not accompanied by the phonoriton spectrum [7] and populates the bottom of the upper polariton dispersion branch.

For negative detunings of the pump light, the STPE signal is proportional to the pump-induced excitonic component at the spectral point $S_2 = [\mathbf{p}_0, \omega_{i=2}(\mathbf{p}_0)]$:

$$I_S \propto \phi^x_{i=2}(\mathbf{p}_0)N_{i=2}(\mathbf{p}_0), \tag{4}$$

where $N_{i=2}(\mathbf{p}_0)$ is the phonoriton occupation number, and $\phi^x_{i=2}(\mathbf{p}_0)$ is the excitonic weight function. The transient population of the LA-phonoriton dispersion branch $i = 2$ is given by $N_{i=2}(\mathbf{p}_0) = \phi^{\rm ph}_{i=2}(\mathbf{p}_0)n^{\rm ph}_{\mathbf{p}_0-\mathbf{k}}$, where $n^{\rm ph}_{\mathbf{p}_0-\mathbf{k}}$ is the LA-phonon occupation number. For $\delta = \overline{S_1S_2} \gg Q_0$, the weight functions are given by $\phi^x_{i=2}(\mathbf{p}_0) = Q_0^2/[(\omega^x_{\mathbf{p}_0} - \omega_{\mathbf{k}} - \Omega^{\rm ph}_{\mathbf{p}_0-\mathbf{k}})^2 + (\Gamma^x)^2]$, $\phi^{\rm ph}_{i=2}(\mathbf{p}_0) = 1 - \phi^x_{i=2}(\mathbf{p}_0) \simeq 1$, and I_S does not depend upon the LA-phonon wave vector $\mathbf{p}_0 - \mathbf{k}$, because $Q_0^2 \propto |\mathbf{p}_0 - \mathbf{k}|$ while $n^{\rm ph}_{\mathbf{p}_0-\mathbf{k}} \simeq k_BT/\hbar\Omega^{\rm ph}_{\mathbf{p}_0-\mathbf{k}} \propto 1/|\mathbf{p}_0 - \mathbf{k}|$.

The maximum of the STPE signal (see Fig. 2) occurs at the resonant condition $\delta = 0$. In this case, however, the pump light already nearly resonates with the exciton level, $\hbar(\omega_t - \omega_{\mathbf{k}}) \simeq 11$ μeV, and effectively populates the orthoexcitonic band. The exciton-exciton scattering becomes dominant, Γ^x increases with intensity, and the phonoriton effect is relaxed. Therefore, for a quantitative analysis of the experimental data by Eq. (4) we will concentrate on negative detunings $\hbar(\omega_{\mathbf{k}} - \omega_t) \le -100$ μeV. For the frequency band specified above, one finds the following using Eq. (4):

$$I_S(\omega_{\rm sig}) \propto \frac{\Omega_c^2(k)I_0}{(\omega^x_{\mathbf{k}} - \omega_{\mathbf{k}})^2} \frac{D_x^2T}{(\omega^x_{\mathbf{p}_0} - \omega_{\rm pr} - \omega_{\rm sig})^2} I_{\rm pr}, \tag{5}$$

where $I_{\rm pr}$ is the intensity of the probe light and $\omega_{\rm pr} + \omega_{\rm sig} = \omega_{\mathbf{k}} + \Omega^{\rm ph}_{\mathbf{p}_0-\mathbf{k}}$. Figure 3 demonstrates that Eq. (5) indeed fits the experimental data on the STPE signal at negative detunings $\hbar(\omega_{\mathbf{k}} - \omega_t) \leq -100\ \mu$eV.

With increasing intensity I_0 of the pump polariton multiphonon scattering processes develop. A *biphoroniton* eigenstate originates from two-LA-phonon-assisted anti-Stokes scattering given by the interaction term $\sum_{\mathbf{p},\mathbf{l},\mathbf{q}}(\tilde{M}_{x-2\rm ph}B^{\dagger}_{\mathbf{p}}B_{\mathbf{l}}c^{\dagger}_{-\mathbf{p}+\mathbf{q}}c^{\dagger}_{\mathbf{l}-\mathbf{q}} + \text{H.c.})$. The biphonoriton is a coherent superposition of two-phonon, exciton, and photon states coupled through the pump polariton $\mathbf{k}$ and characterized by the triplet of the operators $\sum_{\mathbf{l}} c_{\mathbf{p}+\mathbf{l}}c_{\mathbf{l}-\mathbf{k}}$, $B_{\mathbf{p}}$, and $\alpha_{\mathbf{p}}$, respectively. Because of the larger phase space of two-phonon scattering, the pump-induced biphonoriton dispersion has a much less resonant character than that of the phonoriton. We ascribe the nonresonant increase of the STPE signal at pump intensities $I_0 \geq 200$ MW/cm^2 (see the dot-dashed curve in Fig. 2) to the biphonoriton effect.

Because the pump polariton can be interpreted in terms of the laser field induced macro-occupation of the mode $\mathbf{p} = \mathbf{k}$, the phonoriton spectrum, generated by the Bogolubov transformation of the reduced photon-exciton-phonon Hamiltonian (1), can also be attributed to nonequilibrium pump-induced Bose-Einstein (BE) condensation of orthoexcitons. The strongly quantum-statistically degenerate quasiequilibrium distributions of orthoexcitons have been detected in Cu_2O [16]. However, the quadrupole polariton effect prevents the natural BE condensation of orthoexcitons into the mode $\mathbf{p} = 0$ [17]. In contrast, the pump-induced BE condensation of orthoexcitons, which gives rise to the LA-phonoriton spectrum, is detectable, particularly due to an anomalously small absorption coefficient ~ 3 cm^{-1} of the pump light. Recently, kinetics of the laser-induced BE condensation of orthoexcitons were studied in high-precision experiments [18].

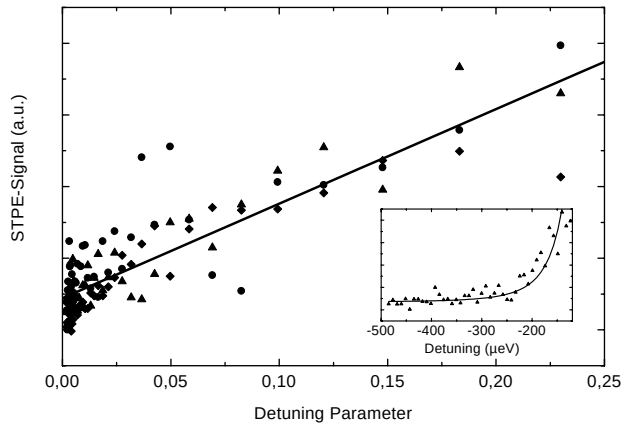

FIG. 3. The normalized phonoriton STPE signal $I_S/I_S^{(0)}$ with $I_S^{(0)} = I_S(\delta = 120\ \mu\text{eV})$ vs detuning parameter $(130\ \mu\text{eV})^4/[(\omega^x_{\mathbf{k}} - \omega_{\mathbf{k}})^2(\omega^x_{\mathbf{p}_0} - \omega_{\mathbf{k}} - \Omega^{\rm ph}_{\mathbf{p}_0-\mathbf{k}})^2]$ for $I_0 = 30$ MW/cm^2 (circles), 45 MW/cm^2 (triangles), and 60 MW/cm^2 (diamonds). According to Eq. (5), all curves coincide and show linear dependence on the detuning parameter. Inset: Fit by Eq. (5) on a linear detuning scale, $I_S = I_S(\delta)$.

In conclusion, we report the first observation of LA phonoritons, composite quasiparticles consisting of orthoexciton, LA phonon, and photon components, which arise in bulk Cu_2O virtually excited by the coherent laser light.

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