

Heterodyne x-ray diffuse scattering from coherent phonons

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Here we report Fourier-transform inelastic x-ray scattering measurements of photoexcited GaAs with embedded ErAs nanoparticles. We observe temporal oscillations in the x-ray scattering intensity which we attribute to inelastic scattering from coherent acoustic phonons. Unlike in thermal equilibrium, where inelastic x-ray scattering is proportional to the phonon occupation, we show that the scattering is proportional to the phonon amplitude for coherent states. The wavevectors of the observed phonons extend beyond the excitation wavevector. The nanoparticles break the discrete translational symmetry of the lattice enabling generation of large wavevector coherent phonons. Elastic scattering of x-ray photons from the nanoparticles provides a reference for heterodyne mixing yielding signal proportional to the phonon amplitude.

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The temperature effect on the x-ray diffraction pattern by crystals reduces the intensity of Bragg peaks by the Debye-Waller factor and produces a weak diffuse inelastic background between the peaks. The leading term of the expansion of this thermal diffuse scattering intensity $I(\mathbf{Q})$ in powers of the atomic displacement u is $\propto \langle u^2 \rangle$ because $\langle u \rangle = 0$, where in this case the expectation value represents a thermal average. In the harmonic approximation u is equally likely to be positive or negative and so $\langle u \rangle^n = 0$ for odd n ¹. This constraint no longer holds for certain non-thermal states, such as coherent phonons².

We recently showed for femtosecond optical excitation of single crystals that the diffuse scattering intensity $I(\mathbf{Q}, t)$ shows temporal oscillations related to the frequencies of phonons corresponding to momentum transfer \mathbf{Q} ³. These oscillations are due to squeezed phonons with $\langle \mathbf{U}_{\lambda, \mathbf{q}} \rangle = 0$ and a time-dependent $\langle \mathbf{U}_{\lambda, \mathbf{q}}^2 \rangle$, where $\mathbf{U}_{\lambda, \mathbf{q}}$ is the phonon displacement at reduced wavevector $\mathbf{q} = \mathbf{Q} - \mathbf{G}$ for the nearest reciprocal lattice vector \mathbf{G} and branch λ ⁴. In contrast, a coherent phonon has time-dependent $\langle \mathbf{U}_{\lambda, \mathbf{q}} \rangle$ as well as $\langle \mathbf{U}_{\lambda, \mathbf{q}}^2 \rangle$ although the mean-squared displacement is constant. In this case, the femtosecond x-ray diffuse scattering intensity acquires a term $\propto \langle \mathbf{U}_{\lambda, \mathbf{q}} \rangle$ that can be comparable to the component at $\langle \mathbf{U}_{\lambda, \mathbf{q}}^2 \rangle$.

For perfect crystals, discrete translational symmetry limits coherent phonon generation to q comparable to the wavevector of the light⁵. Under optical excitation, this limits the generated phonons near the Brillouin zone center ($q \approx 0$). By breaking discrete translational symmetry, this restriction can be modified. For example, in the case of a sharp interface, translational symmetry is broken along the interface normal, and coherent phonons can be generated with non-zero wavevectors perpendicular to the interface⁶. These phonons have been optically generated and detected with time-resolved x-ray diffraction (see for e.g 7–11). In this work, we use nanostructured GaAs (shown in Fig. 1) to break three-dimensional discrete translational symmetry and study the phonon dynamics using time-resolved x-ray diffuse scattering.

We report a femtosecond time-resolved measurement of the diffuse scattering from photo-excited GaAs embedded with ErAs nanoparticles¹² using the Fourier-transform inelastic x-ray scattering (FT-IXS) technique³. The presence of the nanoparticles breaks the discrete translational symmetry of the GaAs host and results in the production of high-wavevector coherent phonons. These manifest in the diffuse scattering as temporal oscillations in $\langle \mathbf{U}_{\lambda, \mathbf{q}} \rangle$, which we distinguish from the more general contribution at $\langle \mathbf{U}_{\lambda, \mathbf{q}}^2 \rangle$ by the time-dependence. The presence of the nanoparticles further served to amplify the signal at $\langle \mathbf{U}_{\lambda, \mathbf{q}} \rangle$ through

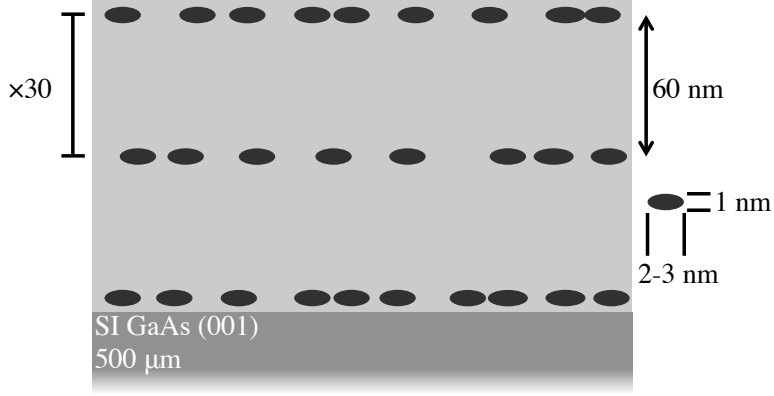


FIG. 1. Cross-section diagram of sample. The black ellipses are ErAs nanoparticles and the gray background is GaAs. See main text for sample details.

the heterodyne mixing between the inelastic scattering from the phonon and the elastic scattering from the nanoparticles.

The measurements were performed at the XPP instrument of the Linac Coherent Light Source¹³. To excite the sample, we used ultrafast 800 nm radiation from a Ti:Sapphire laser system (120 Hz, 20 mJ, 50 fs, p-polarized). The x rays were monochromatic at 8.3 keV with ~ 0.5 eV bandwidth and 10^9 - 10^{10} photons/shot¹⁴. The diffuse scattering measurements were made in a grazing-incidence geometry ($\sim 1.3^\circ$) with the x rays and optical light nearly collinear to match penetration depths. The x-ray spot was $200 \mu\text{m}$ (H) \times $10 \mu\text{m}$ (V) to accommodate the projection along the sample at grazing incidence. All reported measurements were performed using $37.5 \text{ mJ}/\text{cm}^2$ (incident, projected on sample) optical fluence overfilling the x-ray spot. The sample was at room temperature and enclosed in a He environment to diminish low-angle air scattering. The scattering signal was measured using an area detector (CS-PAD¹⁵) on a shot-by-shot basis.

Our sample was a GaAs host with embedded ErAs nanoparticles (referred to below as the nanostructure). As a reference we also looked at pure GaAs. Alternating layers of pure GaAs and GaAs with ErAs nanoparticles were grown epitaxially on a $500 \mu\text{m}$ SI GaAs wafer (surface normal [001]). Each repeated unit was 60 nm of GaAs with 1.2 monolayers of ErAs. There were a total of 30 units ($1.8 \mu\text{m}$ total thickness). We show a diagram of the sample cross-section in Fig. 1. The areal number density of the ErAs nanoparticles was $7 \times 10^{12}/\text{cm}^2$. For details of the sample growth see 12.

Upon excitation of the nanostructure with 800 nm laser pulses, we observed temporal

oscillations in the diffuse scattering intensity. In Fig. 2(A,B), we show two portions of reciprocal space captured simultaneously (that is, for a fixed experimental geometry). Each pixel of the image corresponds to a small volume of reciprocal space, and can be mapped to a specific phonon wavevector \mathbf{q} in the reduced scheme. Panel (A) corresponds to the $(\bar{1}31)$ zone and panel (B) to the $(\bar{1}13)$ zone (conventional unit cell). The ellipses correspond to equal contours of reduced momentum transfer in units of \AA^{-1} (with the convention $q = 2 \sin \theta / \lambda$). The black arrows α and β are paths through reciprocal space which will be discussed below in the context of Fig. 3, while the colored symbols mark particular values of momentum transfer. The change in scattering intensity as a function of time is momentum-dependent, and is illustrated in Fig. 2(C) for the four symbols from Fig. 2(A,B). The value of reduced momentum at each point in units of $\text{m}\text{\AA}^{-1}$ is given in Fig. 2(C). Each time trace shows a few-ps rise time overlaid with rapid oscillations that persist for up to 40 ps. The rapid oscillations are from coherently-excited transverse-acoustic phonons (c.f. 3, 4, and 16) generated by the 800 nm light. As we describe below, these phonons have both squeezed and coherent character.

In Fig. 3, we show the magnitude of the Fast Fourier Transform (FFT) of the time-resolved change in scattering intensity for two paths through the Brillouin zone for both the nanostructure (top panels, labeled ErAs:GaAs) and the GaAs reference (bottom panels, labeled GaAs). The magnitude of the FFT is divided by the static diffuse scattering signal (no optical excitation) and scaled by the square root of the FFT frequency (to better visualize high frequency components). We show data along the black arrows from Fig. 2(A,B). The first path (α) passes close to the Brillouin zone center while the second (β) is further away. The data in the left column are along α while the data in the right column are along β . Note that the two paths (α, β) represent arcs in reciprocal space and thus three coordinates are needed to specify \mathbf{q} . We parameterize these paths with an index indicated on the bottom of Fig. 3. The direction of the arrow in Fig. 2 indicates the direction of increasing index. Along α , we see a clear dispersive feature in both the nanostructure and pure GaAs samples ranging from 1 THz to 2 THz that we attribute to coherently-excited modes in the transverse acoustic branch that modulate $\langle \mathbf{U}_{\lambda, \mathbf{q}}^2 \rangle$ at twice the phonon frequency^{3,4}. There is also a noticeable break in the dispersive feature near path index 15 at approximately 1 THz that indicates the splitting between the two non-degenerate transverse acoustic branches. Similarly along β , we observe intensity ranging from 1.8 to 2.5 THz. This frequency range

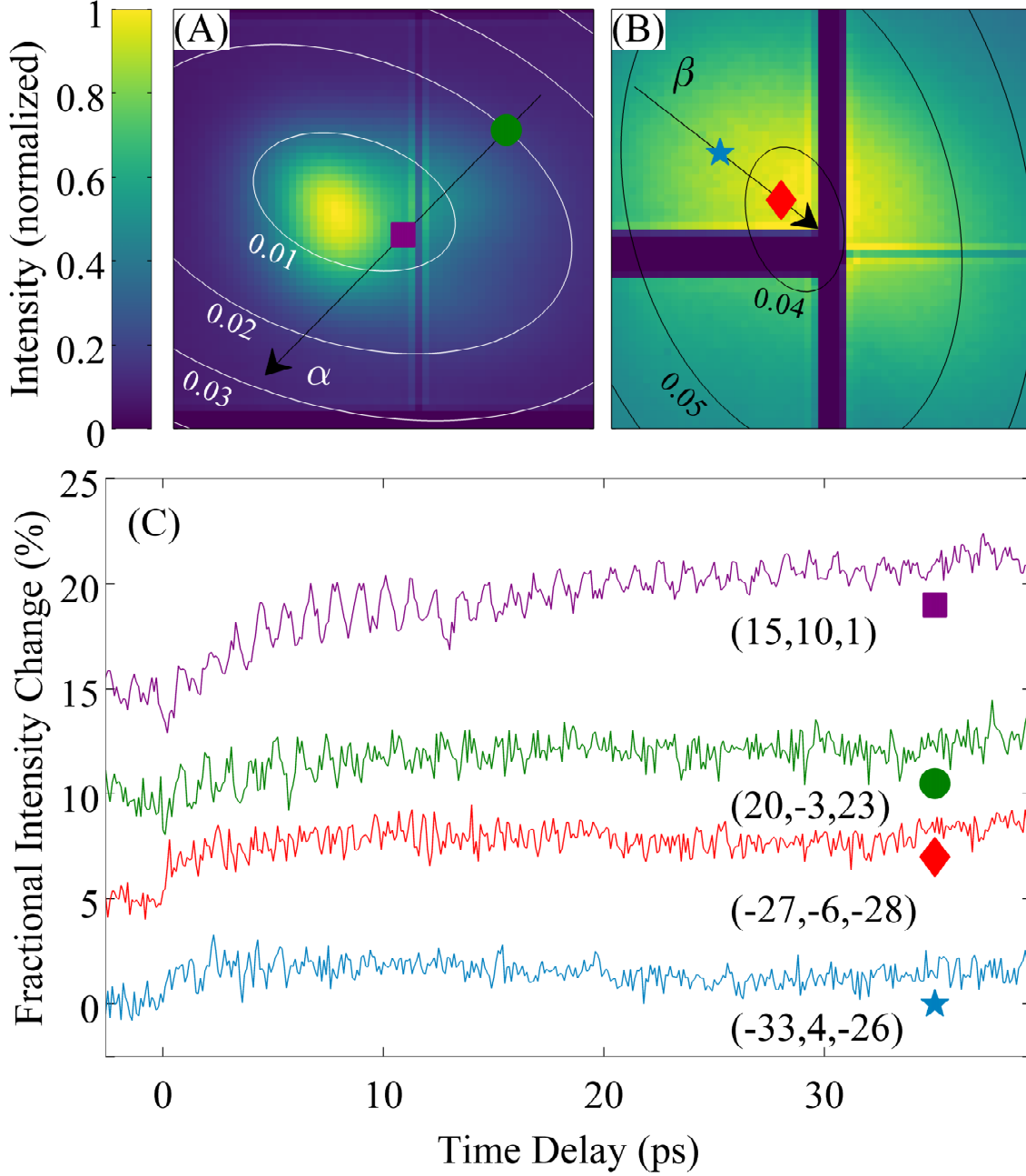


FIG. 2. Diffuse scattering for GaAs with embedded ErAs nanoparticles from the $(\bar{1}31)$ zone (A) and the $(\bar{1}13)$ zone (B). The contours are values of reduced wavevector in \AA^{-1} . The black arrows are paths through reciprocal space considered in Fig. 3. The four shapes (purple square, green circle, red diamond, and blue star) correspond to four points in reciprocal space. The associated dynamics in the diffuse scattering intensity is shown at these four points in (C), along with the value of the reduced momentum at each point in $\text{m}\text{\AA}^{-1}$. Traces are offset vertically for clarity.

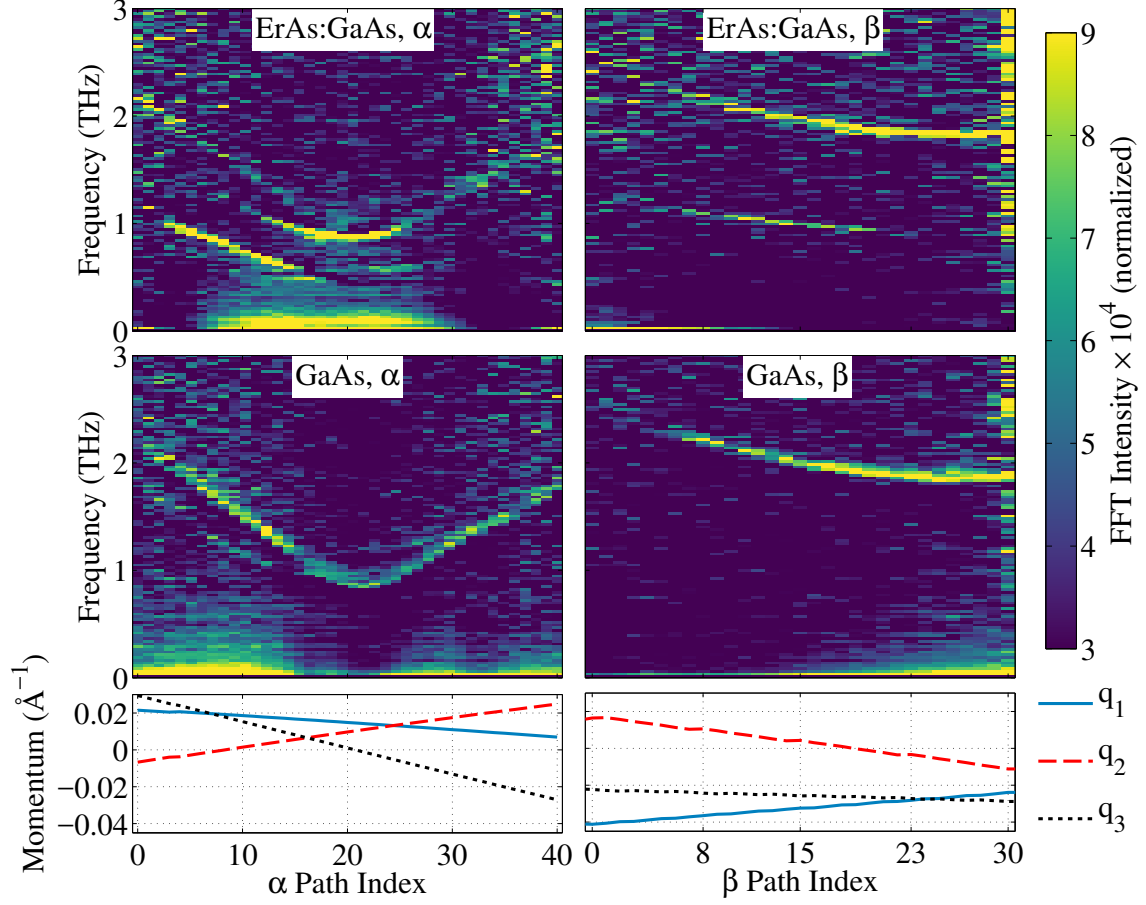


FIG. 3. Frequency-domain analysis of time-resolved diffuse scattering data along black arrows in Fig. 2(A,B) corresponding to specific paths in reciprocal space. The color scale is the magnitude of the Fast Fourier Transform (FFT) divided by the static diffuse scattering signal (no optical excitation) and scaled by the square root of the FFT frequency (to better visualize high frequency components). Each panel is labeled by the material and the path in reciprocal space. The lines below the four panels are the coordinates of the reduced momentum transfer along each path in Fig. 2(A,B) corresponding to the points in the Brillouin zone that are cut by a one-dimensional path in the Ewald sphere. The values on the horizontal axes index the paths in the direction of the arrows in Fig. 2(A,B).

is higher than that along α because the β path is further from the Brillouin zone center.

However, there are additional dispersive features in the nanostructure scattering data that are not present in pure GaAs that we attribute to large-wavevector coherent phonons. Specifically, the data for the nanostructure along α show a dispersive feature from path

indices 3-15 corresponding to the phonon frequency. Along β from indices 5-22 we similarly observe signal at the phonon frequency in the nanostructure. These bands are markedly absent in the GaAs reference sample. We attribute these lower-frequency oscillations to the presence of large-wavevector coherent phonons, enabled by breaking three-dimensional discrete translational symmetry with the nanoparticles.

In order to understand the detection of the additional dispersive bands, we consider the scattered x-ray electric field from the nanostructure. GaAs and ErAs both form on the FCC lattice. For the zinc blende GaAs, in fractional coordinates the As is at the $(0,0,0)$ site and the Ga is at $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$. In ErAs, the structure is rock salt and the As occupies the same $(0,0,0)$ location while the Er is at $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. The epitaxial growth process ensures that the As in the ErAs nanoparticles lie on the same sublattice as the As in the GaAs host^{17,18}. Thus to calculate the total x-ray scattering we account for the presence of the nanoparticles in the GaAs host by replacing clusters of Ga ions with Er, with the appropriate real-space displacement to account for different bases. The scattered field at momentum transfer \mathbf{Q}

$$E(\mathbf{Q}) \propto \sum_{j,s} f_s e^{2\pi i \mathbf{Q} \cdot (\mathbf{R}_j + \boldsymbol{\delta}_s + \mathbf{u}_{j,s})} + \sum_{j'} f_D e^{2\pi i \mathbf{Q} \cdot (\mathbf{R}_{j'} + \boldsymbol{\delta}_{\text{Ga}} + \mathbf{u}_{j',\text{Ga}})}, \quad (1)$$

where f_s , \mathbf{R}_j , $\boldsymbol{\delta}_s$, and $\mathbf{u}_{j,s}$ are the atomic scattering factor (with implicit \mathbf{Q} -dependence), Bravais lattice vector, basis vector, and site-dependent deviation from equilibrium, respectively, for ion s in unit cell j (c.f. 19). The first sum is over all cells j in a perfect GaAs lattice with $s = \text{Ga}, \text{As}$. The second sum is only over unit cells j' where Er has replaced Ga to form nanoparticles. Because the nanoparticles replace Ga for Er, we avoid over-counting the removed Ga from the host by defining an effective scattering factor for the Er,

$$f_D = f_{\text{Er}} e^{2\pi i \mathbf{Q} \cdot (\boldsymbol{\delta}_{\text{Er}} - \boldsymbol{\delta}_{\text{Ga}})} - f_{\text{Ga}}. \quad (2)$$

By assuming that the deviations $\mathbf{u}_{j,\alpha}$ from perfect crystallinity are small, we can approximate each summand to linear order in $\mathbf{u}_{j,s}$. Moreover, we exploit the periodicity of the As sublattice to expand

$$\mathbf{u}_{j,s} = \frac{1}{\sqrt{N}} \sum_{\lambda, \mathbf{q}} \mathbf{U}_{\lambda, \mathbf{q}} e^{2\pi i \mathbf{q} \cdot (\mathbf{R}_j + \boldsymbol{\delta}_s)}, \quad (3)$$

where N is the total number of As sites. The $\mathbf{U}_{\lambda, \mathbf{q}}$ correspond to phonon amplitudes at reduced wavevector \mathbf{q} and branch λ , and have the implicit time-dependence $\exp(-i\omega_{\lambda, \mathbf{q}} t)$

where $\omega_{\lambda,\mathbf{q}}$ is the phonon frequency. Because the perturbations $\mathbf{u}_{j,s}$ are real, $\mathbf{U}_{\lambda,-\mathbf{q}} = \mathbf{U}_{\lambda,\mathbf{q}}^*$. We then substitute this expression into Eqn. 2 and find (see Supplementary Material for detailed calculations)

$$\begin{aligned}
E(\mathbf{Q}) \propto & \mathcal{F}(\mathbf{Q})\mathcal{S}(\mathbf{Q}) + \frac{2\pi i}{\sqrt{N}} \sum_{\lambda,\mathbf{q}} (\mathbf{Q} \cdot \mathbf{U}_{\lambda,\mathbf{q}}) \mathcal{F}(\mathbf{Q} + \mathbf{q}) \mathcal{S}(\mathbf{Q} + \mathbf{q}) \\
& + \mathcal{F}_{\mathcal{N}}(\mathbf{Q})\mathcal{S}_{\mathcal{N}}(\mathbf{Q}) + \frac{2\pi i}{\sqrt{N}} \sum_{\lambda,\mathbf{q}} (\mathbf{Q} \cdot \mathbf{U}_{\lambda,\mathbf{q}}) \mathcal{F}_{\mathcal{N}}(\mathbf{Q} + \mathbf{q}) \mathcal{S}_{\mathcal{N}}(\mathbf{Q} + \mathbf{q}).
\end{aligned} \tag{4}$$

Here $\mathcal{F}(\mathbf{Q})$ and $\mathcal{F}_{\mathcal{N}}(\mathbf{Q})$ correspond to the structure factor for a single GaAs unit cell and an average ErAs nanoparticle treated as a single large unit cell, respectively, at momentum transfer \mathbf{Q} (see Eqn. 6 in the Supplemental Material). We approximate all nanoparticles to be identical in size (1 nm in cross-plane, 2-3 nm in-plane) and orientation (because of the epitaxial growth) but located at random sites throughout the GaAs host^{20–23}. The function $\mathcal{S}(\mathbf{Q})$ is the lattice sum while $\mathcal{S}_{\mathcal{N}}(\mathbf{Q})$ is a truncated version of the lattice sum over only the nanoparticle positions (see Eqn. 6 in the Supplemental Material). Because the nanoparticles are small, $\mathcal{F}_{\mathcal{N}}(\mathbf{Q})$ varies slowly throughout the Brillouin zone. Similarly, $\mathcal{S}_{\mathcal{N}}(\mathbf{Q})$ varies more slowly about the Bragg condition $\mathbf{Q} = \mathbf{G}$ than $\mathcal{S}(\mathbf{Q})$. The first and third terms of Eqn. 4 correspond to elastic scattering at momentum transfer \mathbf{Q} , while the second and fourth terms account for inelastically-scattered components at $\mathbf{Q} + \mathbf{q}$.

To calculate the scattered intensity, it is necessary to multiply Eqn. 4 by its complex conjugate, resulting in several cross-terms. Here we consider only terms linear in $\mathbf{U}_{\lambda,\mathbf{q}}$. These correspond to a heterodyne interference between an elastic component at \mathbf{Q} and an inelastic component at $\mathbf{Q} + \mathbf{q}$, resulting in scattered photons that are offset in frequency by $\omega_{\lambda,\mathbf{q}}$. Guided by one-dimensional simulations (see Supplemental Fig. 1), we expect that the interference term at a particular $\mathbf{Q} + \mathbf{q}$ that has largest contribution is that which mixes the elastic contribution from the nanoparticle at \mathbf{Q} with the inelastic contribution from the host at $\mathbf{Q} + \mathbf{q}$, namely $\Re[\mathcal{S}^*(\mathbf{Q} + \mathbf{q})\mathcal{S}_{\mathcal{N}}(\mathbf{Q})]$. This corresponds to heterodyne detection where the elastic scattering from the nanoparticle acts as the local oscillator. We expect this particular cross-term to be largest because the truncated lattice sum $\mathcal{S}_{\mathcal{N}}(\mathbf{Q})$ for the nanoparticle is much broader in \mathbf{Q} than the full lattice sum $\mathcal{S}(\mathbf{Q})$. Experimentally we directly compare the nanoparticle and host scattering terms by exploiting the difference between the zinc blende (GaAs) and rock salt (ErAs) structures.

In Fig. 4, we show the static diffuse scattering from the nanostructure primarily in the

($\bar{2}22$) zone (conventional unit cell) very close to the zone center. Panel (A) shows the full projection of the zone captured by the detector while (B) shows a zoomed-in region. For rock salt, the ($\bar{2}22$) reflection is strong, while it is weak for zinc blende. Thus, despite the larger volume of GaAs illuminated by the x rays, the scattering signal from the ErAs is much more apparent than in a zone for which both structures have strong contribution (c.f. the ($\bar{1}31$) zone in Fig. 2(A)). As seen in Fig. 4(A) there is non-negligible scattering across a large fraction of the Brillouin zone extending out to 0.025 \AA^{-1} , primarily from the ErAs nanoparticles and directly illustrating the broad variation of $\mathcal{F}_{\mathcal{N}}(\mathbf{Q})\mathcal{S}_{\mathcal{N}}(\mathbf{Q})$. On the other hand, the function $\mathcal{S}(\mathbf{Q} + \mathbf{q}) \sim \delta(\mathbf{G} - \mathbf{Q} - \mathbf{q})$ because of the large number of GaAs unit cells, thus selecting phonons with a well-defined wavevector \mathbf{q} . The ErAs scattering therefore acts as a static background over a large region of the Brillouin zone that can mix with the time-varying GaAs diffuse scattering yielding a heterodyne contribution to the signal. Furthermore we confirm our approximations of a narrow size distribution and orientational arrangement of the nanoparticles by resolving fringes with spatially-varying lobes in the periphery of the scattering (Fig. 4(B)).

In order to observe the heterodyne signal, it is necessary to induce perturbations that enable $\langle \mathbf{U}_{\lambda, \mathbf{q}} \rangle(t) \neq 0$. This is achieved for example via the excitation of a coherent phonon state². Coherent phonon modes cannot be induced at large wavevector in perfectly periodic crystals by optical light because of crystal momentum conservation⁵. The ErAs nanoparticles however break the three-dimensional discrete translational symmetry of the system²⁴, and therefore allow the generation of coherent phonons with wavevectors in all directions. We thus attribute the temporal oscillations at the phonon frequency to coherent phonons observed through the heterodyne detection mechanism. In pure GaAs, however, we only observe temporal oscillations at twice the phonon frequency. Because of long range order, optical radiation cannot generate large wavevector coherent phonons except along the surface normal due to the sharp air/sample interface. However, our measurements do not probe wavevectors along this direction. Thus we attribute the signal in the pure GaAs to squeezed phonons consistent with previous reports^{3,4}.

We have shown that coherent phonons with large wavevector are generated by ultrafast optical excitation in GaAs with embedded ErAs nanoparticles. The nanoparticles break the three-dimensional discrete translational symmetry of the GaAs host and so enable coherent phonon generation at large wavevector in all directions. Moreover x-ray elastic scattering

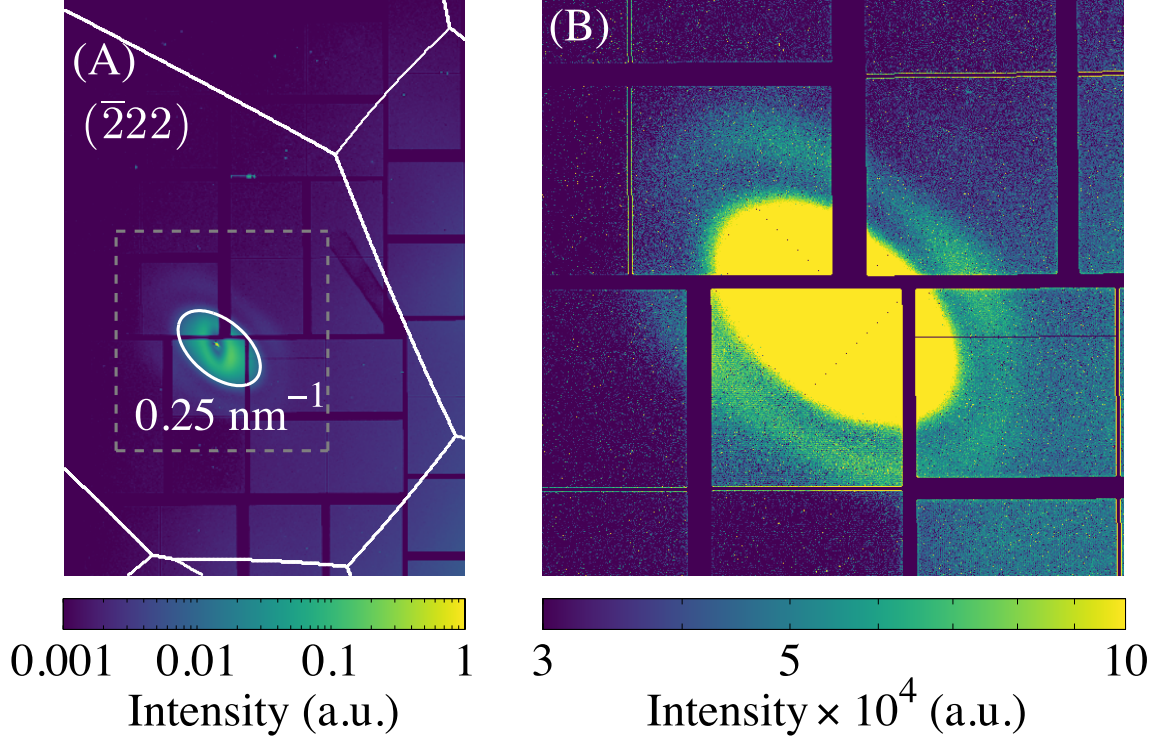


FIG. 4. (A) Static diffuse scattering image of the $(\bar{2}22)$ Brillouin zone for GaAs with embedded ErAs nanoparticles. The white ring corresponds to a contour at 0.025 \AA^{-1} of the reciprocal lattice vector magnitude (reduced zone). The white lines denote Brillouin zone boundaries. The gray dashed box shows the zoom region. (B) Same data as in (A) zoomed in to the center of the $(\bar{2}22)$ Brillouin zone (gray dashed box in (A)) and with a color scale saturated to emphasize the fine features in the periphery.

from the nanostructure interferes with the x-ray inelastic diffuse scattering to provide a heterodyne signal at large wavevector that oscillates at the phonon frequency and is proportional to the amplitude of the phonon mode. Our results show that a small amount of disorder is not detrimental to FT-IXS but in fact can be beneficial due to the signal enhancement from the heterodyne contribution. This represents a first step towards extending FT-IXS for studying vibrational dynamics of disordered materials including alloys and glasses.

SUPPLEMENTARY MATERIAL

See Supplementary Material for the detailed derivation of Eqn. (4) and for the one-dimensional simulation of the nanoparticle scattering.

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