Anisotropic optical properties of detwinned BaFe$_2$As$_2$

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The optical properties of a large, detwinned single crystal of BaFe$_2$As$_2$ have been examined over a wide frequency range above and below the structural and magnetic transition at $T_N \approx 138$ K. Above $T_N$ the real part of the optical conductivity and the two infrared-active lattice modes are almost completely isotropic; only the lattice modes show a weak polarization dependence just above $T_N$. For $T < T_N$, the free-carrier response is anisotropic, being larger along the $a$ axis than the $b$ axis below $\approx 30$ meV; above this energy the optical conductivity is dominated by the interband contributions, which appear to be isotropic. The splitting of the low-energy infrared-active mode below $T_N$ is clearly observed, and the polarization modulation of the new modes may be used to estimate that the crystal is $\approx 80\%$ detwinned. The high-frequency mode, with the threefold increase in strength of the lower branch below $T_N$ and nearly silent upper branch, remains enigmatic.

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I. INTRODUCTION

In the pantheon of iron-based superconductors, the AeFe$_2$As$_2$ (“122”) materials, where $Ae = Ca$, Sr, or Ba, are of particular importance because of the many ways in which superconductivity may be induced [1–15]. At room temperature they are paramagnetic metals, but as the temperature is reduced they undergo a structural transition from a tetragonal ($I4/mmm$) to an orthorhombic ($Fmnm$) unit cell, which is accompanied by a magnetic transition and the formation of spin-density-wave-like (SDW) order where the moments are aligned in the $a$-$b$ planes; ferromagnetically (FM) along the $b$ axis, and antiferromagnetically (AFM) along the $a$ axis. In the case of BaFe$_2$As$_2$, this transition occurs at $T_N \approx 138$ K [16]. In the orthorhombic phase, the crystals are heavily twinned. Early optical studies of the in-plane optical properties of BaFe$_2$As$_2$ examined the average of both orientations in the SDW state [17–19]; however, the application of uniaxial stress along the (110) direction for the tetragonal unit cell results in a nearly twin-free sample [20, 21]. Transport [22, 23], as well as optical measurements [24, 25], of detwinned samples below $T_N$ reveal an anisotropic response where the conductivity along the AFM direction is higher than that along the FM direction; it has been remarked that this is a counterintuitive result as one would normally expect the conductivity to be higher along the FM direction [24]. While the designs of the clamped cells used to mechanically detwin single crystals for optical studies are quite elegant [24, 25], this approach necessarily requires that the size of the imaging spot is smaller than the crystal, leading to a reduced signal and limiting the ability to track weak spectral features such as lattice modes.

In this work we examine the temperature dependence of the optical properties of a large single crystal of BaFe$_2$As$_2$ that has been detwinned through the application of a symmetry-breaking strain, based on differential thermal expansion [26]. While previous studies of detwinned BaFe$_2$As$_2$ have been performed [24, 25], the current study employs overfilling-technique [27] whereby the entire crystal face may be examined, allowing the optical conductivity and the infrared-active modes to be studied. The electronic properties are more or less isotropic for $T > T_N$. The relatively large size of the sample allows the two normally infrared-active modes above $T_N$ to be identified; the lattice modes show a slight polarization dependence just above $T_N$ in response to the applied strain. For $T < T_N$ the free-carrier response is anisotropic, being higher along the $a$ direction, in agreement with previous work. Interestingly, the interband contributions are essentially isotropic. Below $T_N$ the degeneracy of the infrared-active vibrations is lifted and they split into two new modes that are optically-active along either the $a$ or $b$ axis; by examining the polarization modulation of the low-frequency modes below $T_N$, it is possible to estimate that the sample is about 80% detwinned. The behavior of the high-frequency mode is very curious, with one branch increasing dramatically in strength, while the other remains largely silent. This technique for detwining crystals may be used to allow further detailed optical studies of, e.g., the Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ family of materials.

II. EXPERIMENT

Large single crystals of BaFe$_2$As$_2$ were grown by a self-flux method [26]. The crystals have well-defined growth faces, allowing the $a$ axis in the tetragonal phase to be

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identified. A piece of thin glassfiber reinforced plastic (GFRP) was cut and shaped to match the crystal and attached using epoxy with the fibers oriented along the (110) direction in the tetragonal phase, shown in the inset of Fig. 1(a). It has been demonstrated that the difference of the thermal expansion parallel and perpendicular to the fiber direction of the GFRP substrate is comparable to the orthorhombic distortion in BaFe$_2$As$_2$ near $T_N$, resulting in a large symmetry breaking strain; this technique has been used successfully to measure the resistivity and susceptibility anisotropies in BaFe$_2$As$_2$ [26]. The entire arrangement was glued to the tip of an optically-black cone. The temperature dependence of the reflectivity has been measured above and below $T_N$ over a wide frequency range (2 meV to over 3 eV) using an overfilling technique in combination with in situ evaporation [27]; the results are shown in the infrared region in Figs. 1(a) and 1(b) for light polarized along the $a$ and $b$ axes in the orthorhombic phase, respectively. Above $T_N$ the reflectivity along the two polarizations is nearly identical; only the reflectivity at 150 K along the $b$ axis appears to be slightly higher than its counterpart along the $a$ axis. Below $T_N$ the optical properties are strongly anisotropic. A plasma-like edge develops in the reflectivity for both polarizations. For $T \ll T_N$ the low frequency reflectivity approaches unity, while above $\approx 20$ meV the reflectivity decreases rapidly before forming a plateau above $\approx 50$ meV; however, the reflectivity levels and the width of the plasma-like edge are very different along the $a$ and $b$ directions, in agreement with previous optical studies of this material [24, 25, 28]. Additional structure is observed in the mid-infrared region before the reflectivity approaches the values for $T \gtrsim T_N$ above about 0.5 eV (the temperature dependence of the reflectivity is shown over a wide range in Fig. S1 of the Supplementary Material [29]). Superimposed on the reflectance are two sharp features attributed to the infrared-active lattice modes at $\approx 95$ and 256 cm$^{-1}$ [30], which display an anisotropic response below $T_N$. While the reflectance is a useful quantity, it is a combination of the real and imaginary parts of the dielectric function, and as such, is not an intuitive quantity. The complex dielectric function, $\varepsilon(\omega) = \varepsilon_1 + i\varepsilon_2$, has been determined from a Kramers-Kronig analysis of the reflectivity. At low frequency, a metallic Hagen-Rubens extrapolation, $R(\omega) = 1 - A\sqrt{\omega}$ was employed, where $A$ is chosen to match the value of the reflectance at the lowest measured frequency. Above the highest-measured frequency point the reflectance was assumed to be constant to $8 \times 10^4$ cm$^{-1}$, above which a free-electron approximation ($R \propto \omega^{-4}$) was assumed [31].

### III. RESULTS AND DISCUSSION

The temperature-dependence of the real part of the optical conductivity is shown for light polarized along the $a$ and $b$ axis in Figs. 2(a) and 2(b), respectively. Above $T_N$ the real part of the optical conductivity for the two polarizations are almost identical; however, below $T_N$, there is a remarkable anisotropy below $\approx 300$ cm$^{-1}$ where the conductivity along the $a$ direction is larger than along $b$, in agreement with other work [24, 25]. In addition to the broad features associated with the free-carrier response and the interband excitations, there are narrow lattice modes which also display an anisotropic response. The behavior of the electronic properties will be examined first, followed by the lattice modes.

#### A. Electronic response

The optical properties of this multiband material have been studied extensively in the twinned materials and are described by a Drude-Lorentz model where at least two different contributions to the free-carrier response are considered [32], resulting in the complex dielectric
function,
\[
\epsilon(\omega) = \epsilon_\infty - \sum_{j} \frac{\omega_{p,j}^2}{\omega^2 + i\omega/\tau_{D,j}} + \sum_{k} \frac{\Omega_k^2}{\omega_k^2 - \omega^2 - i\omega\gamma_k},
\]

where \(\epsilon_\infty\) is the real part of the dielectric function at high frequency, \(\omega_{p,j}^2 = 4\pi\epsilon_0 e^2/m^*\) and \(1/\tau_{D,j}\) are the plasma frequency and scattering rate for the delocalized (Drude) carriers for the \(j\)th band, respectively; \(\omega_k, \gamma_k\) and \(\Omega_k\) are the position, width, and oscillator strength of the \(k\)th vibration or bound excitation (the intensity is proportional to \(\Omega_k^2\)). The complex conductivity, \(\delta(\omega)\), is calculated from \(\epsilon(\omega)\). The complex dielectric function, \(\delta(\omega) = \sigma_1 + i\sigma_2 = -2\pi\epsilon(\epsilon(\omega) - \epsilon_\infty)/Z_0\), where \(Z_0 \approx 377 \, \Omega\) is the impedance of free space. The real and imaginary parts of the complex conductivity have been fit simultaneously to Eq. (1) using a non-linear least-squares technique.

Above \(T_N\), the reflectivity and the optical conductivity show relatively little polarization dependence. Although the features are rather broad, just above \(T_N\) at 150 K, the fits to the optical conductivity yield a narrow Drude component (D1), \(\omega_{p,D_1} \approx 4100 \, \text{cm}^{-1}\) and \(1/\tau_{D_1} \approx 130 \, \text{cm}^{-1}\), and a broad Drude term (D2), \(\omega_{p,D_2} \approx 11900 \, \text{cm}^{-1}\) and \(1/\tau_{D_2} \approx 1300 \, \text{cm}^{-1}\) (Table I) in good agreement with the values from a previous study [33].

Below the structural and magnetic transition at \(T_N\), the optical conductivity undergoes significant changes, shown in Figs. 1 and 2, due to the reconstruction of the Fermi surface [34]. The fits for \(T \ll T_N\) at 5 K for the \(a\) and \(b\) polarizations are shown in Figs. 3(a) and 3(b), respectively, where they have been decomposed into the individual contributions from the various Drude and Lorentz components. The observed optical anisotropy of \(\sigma_{1,a}/\sigma_{1,b} \approx 2\) in the far-infrared region is in good agreement with another study that employed a mechanical apparatus to detwin the crystal [24]. As in the twinned materials, below \(T_N\) new features appear at \(\approx 350\) and 900 cm\(^{-1}\) [33]; interestingly, there appears to be little or no anisotropy in either these or other bound excitations associated with the interband transitions in this compound. Indeed, the oscillator parameters for the bound excitations in Figs. 3(a) and 3(b) are nearly identical for both polarizations and are similar to what is observed in the twinned material. The anisotropy in the far-infrared region of the optical conductivity arises purely from the behavior of the free carriers. As in the case of the twinned materials, for \(T \ll T_N\) the plasma frequency for the narrow Drude component undergoes only a small decrease, while the scattering rate drops precipitously: along the \(a\) axis, \(\omega_{p,D_1} \approx 3500 \, \text{cm}^{-1}\) and \(1/\tau_{D_1} \approx 3.6 \, \text{cm}^{-1}\); along the \(b\) axis, \(\omega_{p,D_1} \approx 3700 \, \text{cm}^{-1}\) and \(1/\tau_{D_1} \approx 2.1 \, \text{cm}^{-1}\). The uncertainties associated with the small scattering rates, and the similarity of the plasma frequencies, suggest that the narrow Drude component is fairly isotropic below \(T_N\). The plasma frequency for the broad Drude component decreases significantly, while the change in the scattering rate, while significant, is not as dramatic as it is for the narrow Drude component: along the \(a\) axis, \(\omega_{p,D_2} \approx 2900 \, \text{cm}^{-1}\) and \(1/\tau_{D_2} \approx 146 \, \text{cm}^{-1}\); along the \(b\) axis, \(\omega_{p,D_2} \approx 2100 \, \text{cm}^{-1}\) and \(1/\tau_{D_2} \approx 210 \, \text{cm}^{-1}\).
\( \sigma(\omega) (10^3 \Omega^{-1} \text{cm}^{-1}) \)

Data (5 K)
Fit

Figure 3. The results of the fits to the complex conductivity of BaFe\(_2\)As\(_2\) below \( T_N \) at 5 K compared to the real part of the conductivity in the far- and mid-infrared regions for light polarized along the: (a) \( a \) axis; (b) \( b \) axis. The anisotropic response is strongest below \( \simeq 30 \) meV. The fit is decomposed into the contributions from the narrow and broad Drude components, as well as several Lorentz oscillators (Table 1).

The large difference in the plasma frequencies arises from a smaller effective mass along the \( a \) axis where the electronic correlations [35, 36], as opposed to the \( b \) axis, or FM direction, where the larger effective mass significantly reduces the plasma frequency, and subsequent contribution to the optical conductivity in the far-infrared region.

**B. Lattice modes**

In the high-temperature tetragonal phase, the irreducible vibrational representation for the infrared modes yields \( 2A_{2u} + 2E_u \) vibrations [37], where the singly-degenerate \( A_{2u} \) modes are active along the \( c \) axis, and the doubly-degenerate \( E_u \) modes are active in the planes. Below the tetragonal to orthorhombic structural transition at \( T_N \), the degeneracy of the in-plane modes is lifted, \( E_u \to B_{2u} + B_{3u} \), where the \( B_{2u} \) and \( B_{3u} \) modes are active along the \( b \) and \( a \) axes, respectively. In the twinned samples the low-frequency \( E_u \) mode observed at \( \simeq 94 \) cm\(^{-1} \) above \( T_N \) involves the in-plane displacements of the Ba atom moving in opposition to the Fe and As atoms; below \( T_N \) this mode splits into two features at \( \simeq 93 \) and 96 cm\(^{-1} \) [38]. The high-frequency mode at \( \simeq 256 \) cm\(^{-1} \) does not appear to split below \( T_N \), instead displaying an anomalous increase in oscillator strength [18]; this mode involves only the in-plane displacements of Fe and As atoms which move in opposition to one and other [30, 39]. In the detwinned samples, the low-frequency mode has not been examined; the high-frequency mode displays the same anomalous increase in oscillator strength below \( T_N \), but it also does not appear to split and is active only along the \( b \) axis [24]. In this work, we are able to examine the splitting and the polarization dependence of the low-frequency \( E_u \) mode below \( T_N \), and as well as the details of the high-frequency mode.

The low-frequency \( E_u \) mode has been fit using the Lorentzian oscillator described in Eq. (1) superimposed on either a linear or a weak polynomial background. Above \( T_N \) at 295 K, this mode is almost completely isteric, with \( \omega_0 \simeq 93.7, \gamma_0 \simeq 2.5, \) and \( \Omega_0 \simeq 195 \) cm\(^{-1} \).

![Figure 4](image_url)

Figure 4. The real part of the optical conductivity of BaFe\(_2\)As\(_2\) in the region of the low-frequency \( B_{2u} \) and \( B_{3u} \) modes at 5 K for light polarized along the: (a) \( a \) axis; (b) \( b \) axis. For each polarization the vibrational structure has been fit to two Lorentz oscillators superimposed on a linear or weak polynomial background; the fitted oscillator parameters are listed in each panel. The fitted positions of the oscillators are indicated by the arrows.
(Asymmetric line shapes were also considered, but the asymmetry parameter was quite small, effectively resulting in a simple Lorentzian oscillator.) Interestingly, just above $T_N$ at 150 K, there is a slight polarization dependence with $\omega_1 \approx 94 \text{ cm}^{-1}$ along along the $a$ axis, and $\omega_2 \approx 95 \text{ cm}^{-1}$ along the $b$ axis, while the width and strength show no such dependence. This indicates that GFRP is imparting some strain on the crystal and creating a slight asymmetry just above $T_N$. This is consistent with the observation of a significant anisotropy in the resistivity just above $T_N$, which was attributed to the magnetic transition rather than nematic fluctuations [26]. Similarly, the vibrational splitting just above the transition would suggest that the phonons are coupling to the magnetism. Below $T_N$ this vibration clearly splits into two modes at $\approx 94$ and 96 cm$^{-1}$ at low temperature, both of which display a strong polarization dependence.

The optical conductivity is shown in the region of the low-frequency $B_{2u}$ and $B_{3u}$ modes at 5 K for light polarized along the $a$ and $b$ axis, in Figs. 4(a) and 4(b), respectively; the different contributions are denoted in the legend, while the fitted oscillator parameters are shown in the panels of Fig. 4. While the two modes display a strong polarization dependence, the modulation is not perfect. The polarization modulation of the oscillators may be used to estimate the degree to which the crystal is detwinned in the following way,

$$\alpha_j = 1 - \frac{\Omega_j^2(\perp)}{\Omega_j^2(\parallel)},$$

where $\perp$ and $\parallel$ denote the polarizations perpendicular and parallel to the dipole moment of the $j$th vibration. In the case of a twinned crystal, there is no polarization dependence, $\Omega_j(\perp) = \Omega_j(\parallel)$, and $\alpha_j = 0$; if the crystal is completely detwinned, then $\Omega_j(\perp) = 0$ and $\alpha_j = 1$.

Using the parameters for the $B_{2u}$ mode at 5 K yields $\alpha \approx 0.85$; the average for the two polarizations of $\alpha \approx 0.8$ indicates that the crystal is roughly 80% twin free, which is comparable to the estimates based on transport measurements [26].

The high-frequency $E_u$ mode is of considerable interest as one branch displays an anomalous increase in oscillator strength below $T_N$ [18], with the other appears to be largely silent [24]. The high-frequency mode has been fit using a simple Lorentzian oscillator superimposed on a polynomial background; the resulting line shapes are shown with the background removed for light polarized along the $a$ and $b$ axes in Figs. 5(a) and 5(b), respectively. The line shape for this oscillator at 5 K along the $b$ axis has an asymmetric line shape, suggesting electron-phonon coupling; while an asymmetric Fano line shape has been fit to this vibration, the asymmetry parameter is very small [30]. The results for the position, width, and strength of the oscillators along the $a$ and $b$ axes are shown in Figs. 5(c), 5(d), and 5(e), respectively, where they are compared with results from a twinned sample (E $\parallel ab$) [30]. Above 200 K there is no polarization dependence in the oscillator parameters for this mode; however, in a reversal of the behavior observed in the low-frequency $E_u$ mode, just above $T_N$ at 150 K the position of this mode for light polarized along what will become the $a$ axis is slightly larger than it is along the $b$ axis, again indicating that just above $T_N$ the strain from the GFRP is a symmetry-breaking process and that the lattice modes are likely coupling to the magnetism. The frequency of this mode appears to decrease anomalously below $T_N$; this is understood as the splitting of the $E_u$ mode where the lower $B_{2u}$ branch is active and the upper $B_{3u}$ branch is largely silent [30]. The fact that the positions of the modes along the $a$ and $b$ axes are identical [Fig. 5(c)] suggests that the activity of the mode
along the a axis is likely due to leakage from the b axis as a result of residual twins, and that the weak shoulder observed just above this mode at $\simeq 261\,\text{cm}^{-1}$ [Fig. 5(a)] is the $B_{3u}$ mode [38]. The oscillator strength of the $B_{2u}$ mode was observed to increase by a factor of two in the twinned material [18], but in the largely twin-free sample it has increased threefold, in agreement with a previous study [24]. The origin of this anomalous increase in the strength remains a topic of considerable debate. Given the value of $\alpha \simeq 0.8$ determined from the modulation of the low-frequency $E_u$ mode, and the value of $\Omega_0(||) \simeq 340\,\text{cm}^{-1}$ for the $b$ axis at 5 K, then the strength of the leakage should be $\Omega_0(\perp) \simeq 149\,\text{cm}^{-1}$; curiously, the observed value of $\Omega_0(\perp) \simeq 235\,\text{cm}^{-1}$ is considerably larger and suggests a lower value for the detwinning. However, the rather peculiar nature of this mode makes it a poor candidate for estimates of the degree of detwinning. The more predictable behavior of the low-frequency $B_{2u}$ and $B_{3u}$ modes suggests that the estimate of $\simeq 80\%$ detwinning is the more reliable one.

IV. CONCLUSIONS

The optical properties of a large, detwinned single crystal of BaFe$_2$As$_2$ have been determined above and below $T_N \simeq 138\,\text{K}$ over a wide frequency range. Above $T_N$ the optical conductivity and the two infrared-active $E_u$ modes are essentially isotropic; only the lattice modes display a weak polarization dependence just above $T_N$. Below $T_N$, the free-carrier response is strongly anisotropic; below $\simeq 30\,\text{meV}$, $\sigma_{1,a}/\sigma_{1,b} \sim 2$. The narrow Drude component has only a weak polarization dependence. The anisotropy in the low-energy optical conductivity is driven by the difference in the effective masses in the broad Drude component. The interband contributions to the optical conductivity above this energy appear to be isotropic. The relatively large sample size allows the behavior of the lattice modes to be studied in detail. The splitting of the low-energy $E_u$ mode is clearly observed, and the polarization modulation of the resulting $B_{2u}$ and $B_{3u}$ modes are used to determine that the crystal is about 80% detwinned. The high-frequency mode remains enigmatic; the $B_{2u}$ component undergoes a striking threefold increase in intensity, while the $B_{3u}$ mode is nearly silent. This relatively simple method for detwining crystals may allow further detailed optical studies of the nematic and (or) superconducting behavior in this family of materials.

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REFLECTIVITY

The temperature dependence of the reflectivity of a detwinned sample of BaFe$_2$As$_2$ ($T_N \simeq 138$ K) is shown over a wide frequency range for light polarized along the $a$ and $b$ axes in Figs. S1(a) and S1(b), respectively; the axes refer to the orthorhombic phase. The temperature dependent behavior of the reflectivity is restricted to energies below about 1 eV; above this energy there little temperature or polarization dependence. Overall, the spectra are qualitatively similar to those of Nakajima et al. [1].

Figure S1. The temperature dependence of the reflectivity versus the log of the frequency of a detwinned sample of BaFe$_2$As$_2$ for light polarized along the: (a) $a$ axis; (b) $b$ axis.
The temperature dependence of the real part of the optical conductivity determined from a Kramers-Kronig analysis of the reflectivity is shown over a wide range for light polarized along the $a$ and $b$ axes in Figs. S2(a) and S2(b), respectively.

![Graph of optical conductivity vs. frequency](image)

**Figure S2.** The temperature dependence of the real part of the optical conductivity versus frequency of a detwinned sample of BaFe$_2$As$_2$ for light polarized along the: (a) $a$ axis; (b) $b$ axis.

The differences in the optical conductivity in the mid-infrared region appear to be due to the anisotropic behavior of the free-carriers below $T_N$.

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