Inelastic Scattering of Electron Beams by Nonreciprocal Nanotstructures

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Probing optical excitations with high resolution is important for understanding their dynamics and controlling their interaction with other photonic elements. This can be done using state-of-the-art electron microscopes, which provide the means to sample optical excitations with combined meV–sub-nm energy–space resolution. For reciprocal photonic systems, electrons traveling in opposite directions produce identical signals, while this symmetry is broken in nonreciprocal structures. Here, we theoretically investigate this phenomenon by analyzing electron energy-loss spectroscopy (EELS) and cathodoluminescence (CL) in structures consisting of magnetically biased InAs as an instance of gyrotropic nonreciprocal material. We find that the spectral features associated with excitations of InAs films depend on the electron propagation direction in both EELS and CL, and can be tuned by varying the applied magnetic field within a relatively modest sub-tesla regime. The magnetic field modifies the optical field distribution of the sampled resonances, and this in turn produces a direction-dependent coupling to the electron. The present results pave the way to the use of electron microscope spectroscopies to explore the near-field characteristics of nonreciprocal systems with high spatial resolution.

INTRODUCTION

Electron energy-loss spectroscopy (EELS) performed in scanning transmission electron microscopes is a powerful technique to investigate the spatial and spectral characteristics of materials excitations over a wide range of energies [1–10]. More precisely, detailed information on the chemical composition of material structures is routinely gathered by monitoring high-energy losses using this technique [1, 11–13], while the low-loss region of the EELS spectra provides an unsurpassed way of spatially mapping plasmons [8, 14–17], phonons and phonon polaritons [18–22], and excitons [23]. State-of-the-art instruments are currently capable of delivering a combined spectral and spatial resolution in the few-meV and sub-nm range, which allows mapping mid-infrared excitations [17, 18, 20, 22, 24–29]. In addition, the cathodoluminescence (CL) emission associated with electron-driven excitations of optically active modes can equally provide spatially resolved imaging without the requirement of having electron-transparent samples [30–35]. In general, the intensities collected in both EELS and CL depend on the electron trajectory relative to the specimen, and in particular, in virtue of the reciprocity theorem, the EELS probability remains unchanged when reversing the direction of the electron velocity if the sampled structure is made of reciprocal media, while CL is also invariant for systems that possess inversion symmetry along the electron beam (e-beam) direction.

Nonreciprocal photonic structures are currently attracting much attention because they provide appealing ways to control the propagation of electromagnetic waves [36–39]. In particular, gyrotropic materials enable the development of optical circulators based on the Cotton-Mouton effect [40], as well as the realization of one-way electromagnetic wave flow [37, 39] and the manifestation of exotic phenomena such as the violation of detailed balance in the emission of thermal radiation [41]. When exposed to an external DC magnetic field, gyrotropic media display a nonreciprocal response that emanates from the cyclotron orbits described by electrons in the bulk of the material, which result in an anisotropic permittivity tensor with complex off-diagonal components. In a related context, the nonreciprocity arising from a graphene layer that hosts a drift electrical current has been predicted to produce a dependence on the sign of the probe velocity vector in Cherenkov [42] and EELS [43] spectra for free electrons passing near the material with relatively low speeds of the order of the Fermi velocity [42]. Additionally, the presence of an intense external e-beam can trigger nonreciprocal behavior in the guided modes of a metallic cavity [44]. These studies capitalize on the ability of fast electrons to interact with optical modes that do not couple to far-field radiation, but still, a trace of their excitation is directly revealed in the electron-generated spectra. We expect that e-beams can be used to probe the nonreciprocal response with high spatial resolution through the mechanism of near-field coupling to the sample, although this possibility remains so far unexplored.

Here, we predict a large dependence of the EELS and CL spectra on both the strength of an externally applied DC magnetic field and the sign of the probe velocity vector for free electrons interacting with nonreciprocal waveguides that incorporate gyrotropic materials. More precisely, we explore this phenomenon by examining the terahertz spectra associated with planar InAs films, the waveguided modes of which manifest as sharp spectral EELS features undergoing sizeable shifts when
RESULTS AND DISCUSSION

Taking the electron to move with constant velocity $v$ along $z$, following a straight line trajectory determined by $\mathbf{R}_0 = (x_0, y_0)$, the spectrally resolved EELS probability is given by [7, 10]

$$\Gamma_{\text{EELS}}(\omega) = -\frac{4e^2}{\hbar} \int dz \int dz' \text{Im}\left\{e^{i\omega(z'-z)/v} G_{zz}(\mathbf{R}_0, z, \mathbf{R}_0, z', \omega)\right\}$$

in terms of the $zz$ component of the electromagnetic Green tensor $G(r, r', \omega)$, implicitly defined through the relation $\nabla \times \nabla \times G(r, r', \omega) - k^2 \epsilon(r, \omega) \cdot G(r, r', \omega) = (-1/c^2)\delta(r - r')$, where $\epsilon(r, \omega)$ is the position- and frequency-dependent local permittivity tensor, while $k = \omega/c$ is the free-space light wave vector. In structures made of reciprocal materials, the Green tensor satisfies the property $G_{zz}(r, r', \omega) = G_{zz}(r', r, \omega)$, which directly leads to the invariance of $\Gamma_{\text{EELS}}(\omega)$ under the transformation $v \rightarrow -v$ (i.e., reversing the direction of the electron velocity vector). However, in the presence of nonreciprocal materials, the loss probability depends on the sign of $v$.

We consider structures containing indium arsenide (InAs) as an example of material that exhibits a pronounced nonreciprocal response when exposed to a magnetic DC field $\mathbf{B}$ [41]. Using Cartesian coordinates and taking the magnetic field oriented as $\mathbf{B} = By$, the permittivity tensor of InAs becomes [41]

$$\epsilon_{\text{InAs}}(\omega) = \begin{bmatrix} \epsilon_{xx}(\omega) & 0 & \epsilon_{xz}(\omega) \\ 0 & \epsilon_{yy}(\omega) & 0 \\ \epsilon_{xz}(\omega) & 0 & \epsilon_{zz}(\omega) \end{bmatrix},$$

where

$$\epsilon_{xx}(\omega) = \epsilon_{zz}(\omega) = \epsilon_\infty - \frac{\omega_p^2 (\omega + i\gamma)}{\omega (\omega + i\gamma)^2 - \omega_e^2},$$

$$\epsilon_{xz}(\omega) = -\epsilon_{zx}(\omega) = \frac{i\omega_p^2 \omega_e}{\omega (\omega + i\gamma)^2 - \omega_e^2},$$

$$\epsilon_{yy}(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega (\omega + i\gamma)},$$

with $\epsilon_\infty = 12.37$, $\hbar\omega_p = 180.53$ meV, and $\hbar\gamma = 102.02$ meV. The magnetic field enters through the cyclotron frequency $\omega_c = eB/m^*$, where $m^* = 0.33 m_e$ is the effective electron mass in this material. Our structures also contain aluminum (Al) or silicon carbide (SiC), which we describe through their isotropic, frequency-dependent, local permittivities, taken from tabulated optical measurements [46].

For planar structures, we obtain the loss probability by expressing the Green tensor in terms of the Fresnel reflection coefficients $r_{\sigma\sigma'}$, where $\sigma$ and $\sigma'$ run over $p$ and $s$ polarization components. Cross-polarization terms proportional to $r_{sp}$ and $r_{ps}$ emerge as a result of the off-diagonal elements of $\epsilon_{\text{InAs}}(\omega)$ produced in the presence of a magnetic field. Taking the surface at the $x = 0$ plane, we find [47]

$$\Gamma_{\text{EELS}}(\omega) = \frac{e^2 L}{\pi \hbar v} \int \frac{dk_y}{k_p^2} e^{-2k_x x_0} \left( \kappa \text{Im} \{r_{pp} + \frac{k_y^2 \omega^2}{\kappa c^2} \text{Im} \{r_{ss} + \frac{k_y \omega}{c} \text{Re} \{r_{sp} - r_{ps}\}} \right),$$

where $L$ is the length of the electron trajectory, $x_0$ is the electron-surface separation, $\kappa = \sqrt{(\omega/v\gamma)^2 + k_p^2}$ with $\gamma = 1/\sqrt{1 - v^2/c^2}$, and the Fresnel coefficients depend on the loss frequency $\omega$ and the parallel wave vector transfer $k_l = (k_y, k_z)$, and momentum conservation along the beam imposes the condition $k_z = \omega/v$. For lossless dielectric waveguides, the integrand in Eq. (1) vanishes, except at the guided modes, which are signalled by singularities of the Fresnel coefficients. Analytical results for the Fresnel coefficients become too intricate, so we resort instead on numerical simulations in what follows, with the EELS probability $\Gamma_{\text{EELS}}(\omega) = (e/\pi \hbar \omega) \int d\mathbf{r} \text{Re} \{\text{Ext}(\mathbf{r}, \omega) \cdot \mathbf{E}^{\text{ind}}(\mathbf{r}, \omega)\}$ expressed in terms of the self-induced electric field [7] $\mathbf{E}^{\text{ind}}(\mathbf{r}, \omega)$, which is in turn calculated using a frequency-domain finite-difference electromagnetic solver (COMSOL) with the electron source introduced through a line current $\mathbf{j}^{\text{ext}}(\mathbf{r}, \omega) = -e\mathbf{\hat{z}} \delta(x - x_0) \delta(y) e^{i\omega z/v}$. The CL probability is also obtained numerically from the far-field amplitude for the same external source.

Without external magnetic fields, we find four resonance peaks in the EELS spectra simulated from Eq. (1) [black curve in Fig. 1(b)]. The first peak around $12\text{THz}$ (50 meV in energy scale) is associated with the surface plasmon resonances supported at the air-InAs interface, whereas the remaining peaks represent the excitation of plasmon resonances in the InAs film. In the presence of an external magnetic field of strength $B = 1$ T, each of these resonance peaks shifts to the red and blue for electrons traveling along positive (solid blue curve) and negative (dashed blue curve) $z$-directions, respectively, as shown in Fig. 1(b), thus revealing a nonreciprocal response with respect to the electron propagation direction. Incidentally, the electron can be deflected by the magnetic field, but such deflection does not affect the electron spectrum and its magnitude is negligible for electron-sample interaction lengths in the range of a few microns.

To explore the origin of the four resonance peaks observed in Fig. 1(b), we plot the electric field distributions of their associated modes in Fig. 2(a). For the first res-
Aloof electron-beam interaction with a nonreciprocal semiconductor film. (a) We consider electrons moving parallel to an InAs planar film supported on Al and exposed to an in-plane magnetic field $\mathbf{B}$ with the orientation indicated in the figure. The electron-surface interaction depends on the direction of the electron velocity due to the nonreciprocal response induced by the magnetic field in InAs. (b) Simulated EELS probability spectra for electrons moving to the left (dashed curves) or to the right (solid curves) with $B = 1$ T. For simplicity, the results in Fig. 2(b) are obtained by assuming the Al substrate to respond as a perfect electric conductor, although the rest of our calculations take into consideration the metal dielectric function, as indicated above. The waveguide dispersion relations for $k_y = 0$ are then given by the expression

$$\tan(\alpha) = \frac{\epsilon_{zz} \sqrt{\epsilon_1 - \beta^2}}{(\epsilon_{zz} - \beta^2) / \sqrt{\beta^2 - 1} + i \epsilon_{zz} \beta},$$

where $\epsilon_1 = (\epsilon_{zz}^2 + \epsilon_{xx}^2) / \epsilon_{zz}$, $\beta = \kappa z c / \omega$, and $\alpha = (\omega \sqrt{\epsilon_1 - \beta^2 d}) / c$. Clearly, a dependence on the sign of $k_z$ (or equivalently $\beta$) arises in the presence of a magnetic field (i.e., when $\epsilon_{zz} \neq 0$). Incidentally, we can separate the surface mode [Fig. 2(b), red curves], which in the large $d$ limit [i.e., when the modes do not reach the aluminum substrate, as shown in panel (1) of Fig. 2(b)] reduces to

$$\sqrt{\beta^2 - 1} + \frac{\sqrt{\beta^2 - 1}}{\epsilon_1} = -\frac{i \beta \epsilon_{zz}}{\epsilon_1 \epsilon_{zz}}.$$
coupling to emitted light. For electrons traveling along those two opposite directions, light is emitted along different preferential directions derived from umklapp scattering of the guided modes by the grating, so that the corresponding emission angles $\theta_+\text{ and } \theta_-$ (with respect to the positive $x$ axis) satisfy the condition $\theta_+ + \theta_- = \pi$ for $B = 0$, but not in the nonreciprocal structure driven by a finite magnetic field.

CONCLUSIONS

In conclusion, we have demonstrated that free electrons can probe the nonreciprocal response of magnetically biased photonic structures based on gyrotropic materials. We have shown that the observed differences in EELS and CL spectra are apparent for electrons propagating along two opposite directions. We attribute the splitting of multiple waveguide resonance peaks in the electron-generated spectra to the asymmetric dispersions of the applied magnetic field (see color-matched labels).

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FIG. 3: Probing the nonreciprocal response of a buried substrate. (a) We consider an InAs planar substrate coated with a 4.65-μm-thick layer of SiC. Electrons are moving parallel to the film at a distance of 1 μm from the upper surface. An external magnetic field $B$ is applied with the orientation shown in the figure. (b) Simulated EELS probability spectra for electrons moving with velocity $v = 0.5 \, c$ to the right (solid curves) or to the left (broken curves) for different strengths of the applied magnetic field (see color-matched labels).

By patterning a grating on the upper surface of the SiC layer [Fig. 4(a)], waveguide modes are out-coupled to CL emitted light. We calculate the CL signal by integrating the Poynting vector of the emitted light in the far field, from which we observe again a splitting of the waveguide resonance in the CL spectra for electrons moving along opposite directions perpendicular to an externally applied DC magnetic field [Fig. 4(b)]. The corresponding spectral positions of the resonance peaks match very well with those in Fig. 3(b), thus revealing their common physical origin, which is the excitation of waveguide modes whose electric field distributions inside the SiC layer [see Fig. 4(c) for the two peaks labelled in Fig. 4(b)] are very similar to those shown in supplementary Fig. 5 for the planar SiC structure: the grating does not distort these modes substantially, other than to produce out-of-plane electric fields.

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FIG. 4: Nonreciprocal CL emission. (a) We consider a structure similar to that of Fig. 3(a), with a grating carved in the upper surface to produce mode leakage to emitted light. The geometrical parameters indicated in the figure are $P = 10 \mu m$, $W = 5 \mu m$, $d_1 = 0.3 \mu m$, and $d_2 = 4.5 \mu m$. (b,c) Simulated CL probability spectra for electrons moving with velocity $v = 0.5c$ as indicated in (a) either to the right (solid curves) or to the left (broken curves) for different strengths of the applied magnetic field (see color-matched labels) and $1 \mu m$ distance to the top grating surface. The emission is integrated over the full upper hemisphere in (b) and up to angles $< 5^\circ$ relative the $y = 0$ plane in (c). (d) Electric near-field amplitude distributions corresponding to the modes (1) and (2) indicated in (b) for $B = 0$. White lines delineate one period of the SiC grating.
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FIG. 5: (a) Difference between the electric field distributions between the left- and right-propagating split resonance peaks shown in Fig. 1(b) (see corresponding labels there) with an applied DC magnetic field of strength $B = 1$ T. (b) Same as Fig. 4(c), but in the presence of a magnetic field of strength $B = 0.3$ T.

FIG. 6: Calculated EELS probability for the same configuration as depicted in Fig. 1(a), but for different electron velocities: (a) $v = 0.67c$ and (b) $v = 0.4c$. 