Terahertz Tuning of Dirac Plasmons in Bi_2Se_3 Topological Insulator

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Light can be strongly confined in subwavelength spatial regions through the interaction with plasmons, the collective electronic modes appearing in metals and semiconductors. This confinement, which is particularly important in the terahertz spectral region, amplifies light-matter interaction and provides a powerful mechanism for efficiently generating nonlinear optical phenomena. These effects are particularly relevant in graphene and topological insulators, where massless Dirac fermions show a naturally nonlinear optical behavior in the terahertz range. The strong interaction scenario has been considered so far from the point of view of light. In this Letter, we investigate instead the effect of strong interaction on the plasmon itself. In particular, we will show that Dirac plasmons in Bi_2Se_3 topological insulator are strongly renormalized when excited by high-intensity terahertz radiation by displaying a huge red-shift down to 60% of its characteristic frequency. This opens the road towards tunable terahertz nonlinear optical devices based on topological insulators.

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Nonlinear phenomena play a fundamental role in modern photonics, by enabling many optical functionalities like ultrashort pulse generation and shaping, sum- and difference-frequency processes and ultrafast switching. These effects usually happen in a host material and can be described in terms of an effective photon-photon interaction governed by the material's properties. However, most of nonlinear optical effects are extremely weak and have been observed only in the visible and near-infrared spectral regions. In order to enhance the local electromagnetic field and extending nonlinear phenomena towards the technological important terahertz (THz) spectral range, many strategies have been proposed. These proceed along two main directions: the investigation of novel nonlinear materials, for instance graphene [1,2], topological insulators (TI) [3], and 3D Dirac systems [4] whose low-energy electrodynamics is characterized by massless Dirac fermions having an intrinsically nonlinear THz response [5,6], and material engineering through cutting edge nanotechnologies [7]. A strong nonlinear THz response can be obtained by combining these two strategies.

Topological insulators are quantum materials exhibiting an insulating electronic gap in the bulk, whose opening is due to strong spin-orbit interaction, and gapless surface states at their interfaces [9]. These surface states are metallic, characterized by a Dirac dispersion, showing a chiral spin texture [10,11] and protected from backscattering by the time-reversal symmetry. Furthermore, TI surface states spontaneously provide a 2D Dirac-fermion system, segregated from the bulk material without the need of physically implementing an atomic monolayer.

The optical properties of TIs are characterized by a free-particle Drude term in the THz region superimposed to a phonon absorption. The Drude term is mainly due to Dirac carriers [12] which also substain electronic collective excitations, i.e., Dirac plasmons [12,13].

Plasmons provide the unique possibility to store in subwavelength spatial regions the information encoded in a light beam, a property which is of the highest interest for nano-electronics applications and already exploited in nano-(bio) sensors for the detection of vanishingly small quantities of a given analyte [14,15]. The subwavelength confinement implies that the electromagnetic energy associated with the light field is highly concentrated in so-called hot spots, where the interaction of light and matter is strongly enhanced thus allowing for ultrafast processing of optical signals down to the femtosecond timescale. This strong interaction scenario has been considered so far mainly from the point of view of light, with the plasmon hot spots providing an efficient transduction mechanism to induce high-order light-mixing processes and enabling new optical functionalities [16]. The study of the nonlinear effects on the plasmon itself, like changes on its characteristic excitation frequency due to the strong (nonlinear) interaction with light, have been much less addressed so far from an experimental viewpoint.

In this Letter, we fill this gap by studying the plasmonic response in the terahertz region in Bi_2Se_3 TI thin films when Dirac plasmons are excited by a strong THz electric field in the MV/cm range. Bi_2Se_3 TI molecular beam epitaxy (MBE)-based thin films are particularly suitable for optical measurement. Indeed, through the MBE growing technique, one minimizes defects and out-of-stoichiometry, strongly reducing extrinsic optical absorption from the bulk and obtaining a nearly pure Dirac response from surface [3,12,17]. A THz plasmonic excitation can be achieved if the TI surface is patterned, for instance in the form of parallel ribbons, thereby providing to THz light the necessary momentum to match, in the absorption process, the plasmon's dispersion relation (see Fig. 1). Dirac-based THz plasmons have been indeed observed in the past few years, both in graphene [18] and TIs [13,19–23]. We show that Dirac plasmon resonances in Bi_2Se_3 ribbon array films are strongly renormalized upon intense THz illumination, by displaying a red-shift down to 60% of their linear value measured at low E_{THz} field. Our observation proves that TI nanoribbons provide a viable way towards a *light-only* active ultrafast plasmon manipulation and tunable sensors.

Results.—We have prepared two different patterned films of Bi_2Se_3 in form of parallel ribbons with a period $2w$ and widths $w = 4$ and 20 μm (filling factor 1/2). The THz transmittance $T(\nu)$ in the linear regime were measured with the synchrotron-based FTIR setup of the SISSI beam line [24] at the Elettra light source, providing THz pulses at 500 MHz repetition rate with an associated electric field of about 0.1 kV/cm. In order to investigate a possible THz induced nonlinear plasmon behavior, we have employed the TeraFERMI [8,25] source at FERMI@Elettra Free-Electron Laser, providing high-power broadband sub-ps THz pulses, coupled with a homemade step-scan Michelson interferometer.

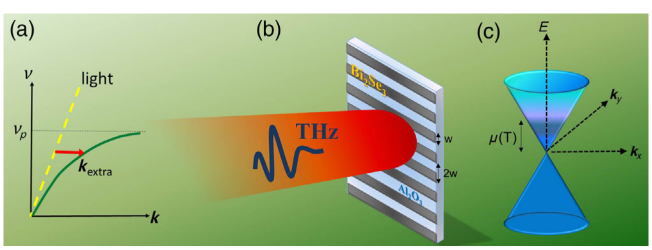

FIG. 1. Plasmons in a topological insulator. Plasmons are collective oscillations of electrons that can be directly excited by electromagnetic radiation in the presence of an extra momentum k_{extra}. This is achieved in the present experiment (a),(b), through ribbon arrays (width w, period $2w$, and $k_{extra} = \pi/w$), fabricated onto the surface of topological insulator Bi_2Se_3 films (a) with linearly dispersive electronic bands (c). Plasmons are excited after illumination with sub-ps, half-cycle THz pulses produced at the FERMI free-electron laser [8].

The extinction coefficient $E(\nu) = 1 - T(\nu)$, as extracted from transmittance $T(\nu)$ measurements with light polarized perpendicular to the ribbons, is reported in Fig. 2 for the two films.

In the linear regime (i.e., for $E_{THz} = 0.1$ kV/cm) one identifies a two hump structure (see Fig. 2) in the extinction spectrum of the Bi_2Se_3 ribbon array patterned films. As already shown in Ref. [13], this peculiar absorption feature can be attributed to the THz plasmon resonance which is activated by the patterned structure, coupled via a Fano interaction, with the α phonon at nearly 2 THz. In particular, the low energy peak observable in the $E(\nu)$ spectra (between nearly 0.8–1.5 THz in Fig. 2) corresponds to the dressed plasmon, while the high energy mode (between nearly 2–2.5 THz) to the dressed α phonon. A closer look to the raw extinction data in Fig. 2 (empty circles) and in particular to the low-frequency maxima shows an evident softening [arrows in Figs. 2(a)–2(e) and 2(f)–2(i)] of the dressed plasmon mode.

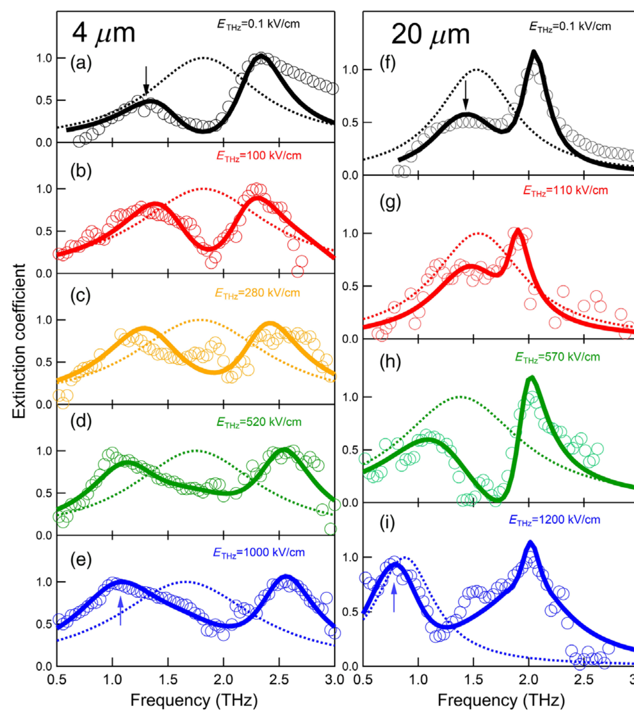

FIG. 2. Fluence-dependent THz spectra. Panels (a)–(e) display the THz extinction coefficient $E(\nu)$ of the $w = 4$ μm ribbon array patterned film for different THz electric field values. THz light is perpendicularly polarized to the ribbons. $E(\nu)$ shows a plasmon-phonon coupled spectrum (see main text). In particular the low-frequency maxima correspond to the dressed plasmon and the high-frequency modes to the dressed α phonon. The full line is a fit according to Eq. (1). The same is shown in panels (f)–(i) for the $w = 20$ μm patterned film. Both the dressed plasmons (see arrows) and the bare plasmon absorption [dashed lines as calculated from Eq. (1)], show an evident softening vs the THz electric field.

In order to map the softening of the dressed plasmon to the corresponding softening of the bare plasmon, we fit the extinction coefficient through the following equation obtained by Giannini *et al.* [26]:

$$E(\nu') = \frac{[\nu' + q(\nu')]^2}{\nu'^2 + 1} \times \frac{g^2}{1 + (\frac{\nu - \nu_p}{\Gamma_p/2})^2}, \qquad (1)$$

where ν_p (Γ_p) is the bare plasmon frequency (linewidth) and ν_{ph} is the bare phonon frequency. The expressions for the renormalized frequency ν' and for the Fano parameter q, that is the ratio between the probability amplitude of exciting a discrete state (phonon) or a continuum or quasicontinuum state (plasmon), are reported in the Supplemental Material [27]. Finally g is the plasmon-radiation coupling factor.

After fitting data from Fig. 2, to Eq. (1), we can establish the THz electric field dependence of the bare plasmon frequency ν_p, as reported in Fig. 3 (see the Supplemental Material [27] for the E_{THz} values estimate). With increasing field the bare plasmon frequency of the 4 μm-ribbon film shifts from 1.8 at 0.1 kV/cm (the linear regime value) to 1.6 THz at the highest field (∼1 MV/cm), i.e., 85% of its linear value. Analogous results are also found for the film with 20 μm-ribbon pattern for which the field-dependent bare plasmon frequency changes from 1.5 to 0.9 THz, thus resulting in a plasmon softening of about 60%. A similar softening is shown by the dressed plasmons for both 4 and 20 μm ribbon arrays, when their frequencies are extracted from extinction data through a Lorentz fitting (see Fig. S1 in [27]). Let us observe that the THz-induced plasmon-softening is at variance with optical pump-THz probe results. Here, indeed, a plasmon hardening has been observed [32] due to an effective increase of the Dirac carrier population determined by the pumping above the optical gap of Bi_2Se_3.

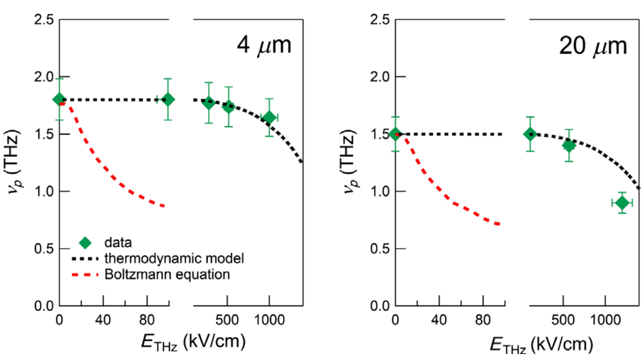

FIG. 3. Plasmon softening. Experimental plasmon frequency ν_p values for both patterned films are shown here as a function of the THz electric field E_{THz}. According to the Boltzmann equation formalism a softening of the plasmon (dashed red line) is theoretically predicted [6]. This model fails in reproducing the experimental data. On the other hand, a thermodynamic model (black dotted line), correctly catches our experimental observations (see main text).

Discussion.—A softening of the plasmon frequency at high THz fields was recently predicted in graphene by several authors [6,33]. One theoretical approach to the problem consists in solving the Boltzmann equation of transport in a nonperturbative way [5,6]. The nonlinear properties can be completely defined by one dimensionless parameter $\mathcal{F}_\tau = \tau e E_{\mathrm{THz}}/\hbar k_F$, where e is the electron charge, E_{THz}, the THz electric field, τ the scattering rate of free Dirac electrons, and k_F the Fermi wave vector. $\mathcal{F}_\tau$ essentially measures the ratio of the energy transferred by the THz pulse to a single electron between two scattering events with respect to the Fermi energy (see the Supplemental Material [27]). The model predicts a very significant plasmon softening at relatively low fields (∼10 kV/cm) which is however not found in our experimental data (Fig. 3, red dashed line). A probable explanation for this disagreement is related to the low heat capacity associated to the massless Dirac electrons. Indeed, the temperature of the electron bath undergoes a strong enhancement upon intense THz excitation (which is actually not considered in the Boltzmann equation model), thus resulting in a nonlinear response dominated by incoherent thermal effects [33].

In order to take into account the local variation of temperature due to the absorption of the THz energy, we adopt a standard two-temperature model [34,35]. Here, the electronic temperature T_e of our Bi_2Se_3 films can be determined by numerically solving the coupled differential equations:

$$c_e(T_e)\frac{dT_e}{dt} = -G(T_e)(T_e - T_l) + P(t), \qquad (2)$$

$$c_l(T_e)\frac{dT_L}{dt} = G(T_e)(T_e - T_l), \qquad (3)$$

where T_e and T_l are the electronic and lattice temperatures respectively, $c_e(T_e)$ is the electronic specific heat, $G(T_e)$ describes the coupling between the electron and phonon subsystems, $c_l(T_e)$ is the lattice specific heat, and $P(t)$ is the THz instantaneous power absorbed by the film. As detailed in the Supplemental Material [27], all these parameters are available from literature. As a consequence of the THz pulse absorption, the electronic temperature of Bi_2Se_3 reaches up to 1500 K for fields of about 1 MV/cm. This value is significantly lower than the Fermi temperature in Bi_2Se_3. Let us notice that T_e in Bi_2Se_3 is also lower than in graphene, where, due to a less effective interaction of electrons with the lattice, electronic temperatures as high as 6000 K are observed under the same illumination conditions [36]. Similarly to graphene, the rise of T_e follows the pump profile $P(t)$ within few 10s of fs, and relaxes back to the lattice temperature within few ps [27].

The increase of the electronic temperature due to the THz illumination can be traced in a strong modification of the electronic chemical potential and, finally, in the softening of the plasmon frequency. Indeed, in a Dirac system the Sommerfeld expansion of the chemical potential at the first order writes $\mu(T_e) \sim \epsilon_F[1 - (\pi^2 k_B^2 T_e^2/6\epsilon_F^2)]$ [36,37], which directly mirrors in the temperature dependence of $\nu_p \propto \mu(T_e)^{1/2}$.

According to a pure thermodynamic model, this relationship can be used to find the dependence of ν_p on E_{THz} which is shown in Fig. 3 (black dotted line). The predicted plasmon frequencies, are in remarkable agreement with the experimental data, with a minor deviation observed only at the 1.2 MV/cm THz field point for the 20 μm patterned film. This deviation could be due to a thermal excitation of electrons from the surface into the bulk, inducing a further softening of plasmon not considered in Eqs. (2) and (3). The thermodynamic model has been already used to describe the field-dependent THz nonlinear properties of graphene, like its optical conductivity [1], harmonics generation [2], and pump-probe dynamics in ribbon arrays [36,37]. It is also known that when the electronic temperature increases, the electron-electron scattering rate increases as well. Since the Boltzman kinetic equation does not take into account electron-electron many-body effects [38], its applicability for high THz pump intensities becomes questionable, while a hydrodynamic regime uniquely characterized by frictional forces between electrons may eventually apply for high enough T_e [39]. In the case of TIs, a purely hydrodynamic regime is probably hampered by the presence of electron-phonon scattering channels which are much more significants than in graphene. However the increase in T_e is still high enough to make the Boltzmann kinetic approach not suitable to the description of our data.

We have shown here, that Dirac plasmons in Bi_2Se_3 topological insulator ribbon arrays can be tuned with THz light, over a wavelength range spanning almost one octave. In the regime explored in this Letter, the plasmon softening effects can not be described with a conventional Boltzmann equation model, while a thermodynamic picture, similar to that recently applied to graphene, captures most of our findings. The strong renormalization of plasmon excitation observed here, is extremely promising for the exploitation of topological insulators as a platform for the realization of active plasmonic devices, their unique properties in terms of spin-polarized currents representing an additional asset for ultrafast nanospintronics applications.

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