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ABSTRACT

The cleaving of a solid to form two new surfaces may result in the emission of light. Conventional mechanoluminescence involves the transfer of charge *between* the two surfaces. We now demonstrate that the ultra-fast separation of charge *within* a newly-formed surface will lead to the emission of electromagnetic radiation. In contrast to the visible light previously observed and modeled, the intra-surface radiation contains terahertz frequencies. This new mechanism – named here surfoluminescence – introduces a new class of terahertz-frequency emitters. It also may in part explain the recent observation of terahertz emission from peeling adhesive tape.

1. Introduction

It has been long known that visible radiation may be produced by mechanical force. This phenomenon is known as mechanoluminescence. Specific examples include fractoluminescence, the emission of light from breaking surfaces, such as when sugar is crushed [1], and triboluminescence, the emission of light associated with friction, such as when adhesive tape is peeled from a surface [2,3]. While the mechanisms are not understood in detail, it is generally accepted that the luminescence depends on the separation of charge between two surfaces. Here we show that the creation of a surface itself, as distinct from the interaction between two surfaces, is sufficient to produce electromagnetic radiation.

While the early observations of mechanoluminescence were restricted to visible light, more recent experiments have shown that a wider spectrum of electromagnetic radiation may be emitted. For example, peeling adhesive tape in a vacuum produces X-ray [4] as well as radio-frequency [5] radiation. It has recently been reported that peeling adhesive tape also emits terahertz-frequency electromagnetic radiation [6]. This appears to be the first report of terahertz emission related to mechanoluminescence.

We here show that the formation of a surface, in the presence of a surface potential and mobile charge carriers, leads to a transient charge distribution that forms a radiating dipole. The charge redistribution occurs on the timescale of a picosecond, resulting in electromagnetic radiation on the frequency scale of the terahertz. We model the process using an ensemble Monte Carlo simulation. As a particular example we give detailed results for the compound semiconductor GaAs as a function of surface potential and doping concentration.

We emphasize that the mechanism proposed here differs from conventional mechanoluminescence in a number of key aspects. First, the transfer of charge is within, rather than between, surfaces. Secondly, the radiation emitted is at terahertz, rather than at visible (or X-ray, or radio) frequencies. Thirdly, the mechanism is the one-step process of the formation of a rapidly-changing dipole, rather than the multi-step process of charge separation, leading to electrical breakdown, with subsequent ionization of the surrounding atmosphere, then possible involvement of bremsstrahlung. Fourthly, only mobile charge carriers of one sign, rather than both anions and cations, are required. Fifthly, the mechanism only requires the formation of a new surface, whether this be by the cleavage or crushing associated with fractoluminescence, the peeling or friction associated with triboluminescence, or rubbing, scratching, sonification or other processes associated with a generic mechanoluminescence. Lastly, the strength of the emitted signal depends on the material properties of surface potential and charge-carrier concentration, increasing with each of these in a systematic way. In view of these distinctive features, we suggest the specific name 'surfoluminescence' for the new mechanism proposed here.

2. Model

A three-dimensional ensemble Monte-Carlo numerical simulation is used to investigate the carrier dynamics in a semi-classical way [7]. We solve the electric potential self-consistently with the charge distribution using a finite-difference method. Phonon scattering and interband scattering are taken into account. Between scattering events, the charge-carrier transport is ballistic. The electromagnetic radiation is then calculated from the time derivative of current density. This is broadly the same general method used by a number of others in accounting for the terahertz radiation produced by ultrafast

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optical excitation of semiconductor surfaces [8–16]. Monte Carlo methods have superseded the analytical approach to surface generation of terahertz radiation, a point illustrated by the earlier analytical [17] and later Monte Carlo [13] modeling of Malevich.

We solve the equations for electron and hole motion

$$\ddot{\mathbf{r}} = \frac{q}{m}\mathbf{E}(\mathbf{r}, t) \tag{1}$$

using Euler's method. Here $\mathbf{r}$, q, and m are respectively the position, charge, and mass of the charge carrier. After every time interval Δt the electric field $\mathbf{E}$ is recalculated using the finite differencing approach for a cubic grid that leads to a set of simultaneous equations, $\phi_{x,y,z} =$

$$\frac{1}{6}\left(\phi_{x+1,y,z} + \phi_{x-1,y,z} + \phi_{x,y+1,z} + \phi_{x,y-1,z} + \phi_{x,y,z+1} + \phi_{x,y,z-1} + \frac{\rho_{x,y,z}(\Delta x)^2}{\epsilon_0 \epsilon_r}\right). \tag{2}$$

These equations are solved numerically by an successive-over-relaxation (SOR) method [18] and the resulting fields used to update the velocity and position of each particle. Each face of each elementary cell is subject to boundary conditions. The potential at the semiconductor surface is fixed (Dirichlet condition) at the surface potential arising from band bending. The four elementary cell surfaces perpendicular to this have Dirichlet values fixed by the adjacent cells; the outermost cells have cyclic values. The final face has a constant spatial derivative of potential, $\partial\phi_{x,y,z}/\partial x = 0$ (Neumann condition). This implies that the electric field $\mathbf{E} = -\nabla\phi$ vanishes gradually into the sample.

Energy loss and direction change on scattering are calculated for several mechanisms. The total scattering rate is the sum of the polar-optical phonon, intervalley, and ionized impurity scattering in the Conwell and Weisskop approach [7]. Polar-optical phonon scattering occurs at a rate

$$P(\epsilon) = \frac{e^2\sqrt{m}\omega_{op}}{\sqrt{2}\hbar}\left(\frac{1}{\kappa_\infty} + \frac{1}{\kappa_0}\right)\ln\left(\frac{\epsilon^{1/2} + \epsilon'^{1/2}}{\epsilon^{1/2} - \epsilon'^{1/2}}\right)\frac{f_{BE}\left(\hbar\omega_{op}\right)}{\epsilon^{1/2}}. \tag{3}$$

The polar-optical phonon rate with scattering angle is

$$P(\theta) = \frac{\epsilon\epsilon' \sin(\theta) d\theta}{\epsilon + \epsilon' - 2\sqrt{\epsilon\epsilon'}\cos(\theta)}. \tag{4}$$

The intervalley scattering is taken to be isotropic, with the scattering rate given by

$$P(\epsilon) = \frac{(D_t K)^2 m^{3/2} Z_f}{\sqrt{2}\pi\rho\hbar^3\omega_i}(\epsilon \pm \hbar\omega_i - \Delta\epsilon_{fi})^{1/2} f_{BE}(\hbar\omega_i) \tag{5}$$

and the ionized impurity scattering is also considered in the Conwell and Weisskop formulation:

$$P(\epsilon) = \pi n_i Z^2 \left[\frac{3}{4\pi n_i}\right]^{1/3}\sqrt{2\epsilon/m}. \tag{6}$$

The density of electrons or holes is determined by the intrinsic and extrinsic doping level. We assume that the temperature is sufficiently high for the given material that all doped sites are thermally ionized. Initially all mobile electrons and holes are positioned randomly. The uniformly-distributed immobile charge at the donor or acceptor ions is also included in the calculation. The simulation is then run until equilibrium occurs. A (near) zero net current indicates that the equilibrium distribution has been reached. The charge thus reconfigures itself self-consistently with the surface field yielding a depletion or accumulation layer. The emitted radiation in the far-field approximation is taken to be proportional to the rate of change of current.

3. Results and discussion

Fig. 1 shows the result of running the simulation for a GaAs crystal of electron doping $5\times10^{15}\,\mathrm{cm}^{-3}$. The potential begins at zero throughout the whole volume before the surface is created at $t = 0$. Immediately there is a strong suppression of the potential at the surface. This extends more than 1 μm. Shortly after, shown at 1, 2, 3, and 4 ps after the surface formation, a strong oscillation in the potential is seen extending to the depth of several μm. The origin of the oscillation is in the transient redistribution of the charge carriers. It is this oscillation which is the physical basis of the emission of electromagnetic radiation of terahertz frequencies. After a short time (15 ps shown) the transient effects have subsided.

Our principal results are presented in Figs. 2 and 3. The vertical axes are in normalized units defined previously for a standard case of InAs [20,21]. Both figures show the radiation in the far field that results from the carrier dynamics that give rise to the potential dynamics illustrated in Fig. 1.

Fig. 2 demonstrates that, as expected, a higher surface potential gives rise to a higher emission of electromagnetic radiation. With the parameters employed here, the increase in radiated field is almost linear with the increase in surface potential. This relation would give rise to a quadratic dependence of radiated power on surface potential.

We have calculated in detail the effect for small surface potentials, for example, 0.1 V for InAs, and found negligible emission of terahertz radiation in this case [20]. Our further calculations on attractive (positive) surface potentials show emission is possible if the potential is high enough. However, there are only very weak secondary oscillations because the electrons are forced to continuously recollide with the surface. This leads to a random phase, and highly noisy variations in the field and charge distributions.

Fig. 3 shows the variation in the emission in the far field with doping. As expected, the emission increases with doping level. The dependence is approximately linear.

In comparing Figs. 2 and 3, it is seen that the period of the pulse and of the secondary oscillation is affected by the doping level and is comparable to the bulk plasma frequency, but is not strongly affected by the surface potential.

We approach the question of the possibility of detecting terahertz surfoluminescence from two perspectives. First, we compare the electric field we calculate on surface creation with the electric field we calculate for optical excitation under typical laboratory conditions (1 nJ laser strike of 790 nm radiation of pulse length 10 fs). As may

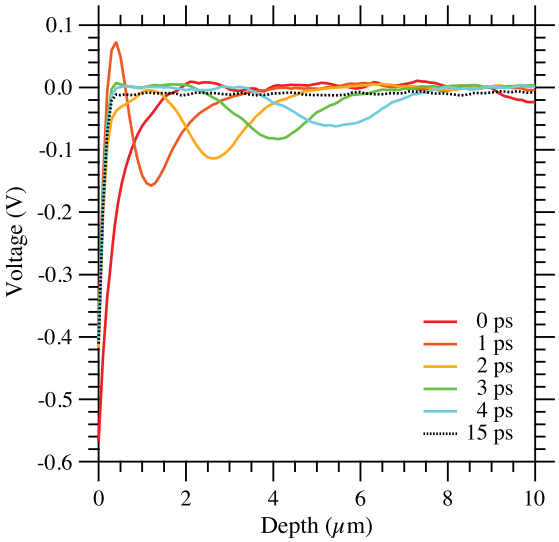

Fig. 1. Electrical potential as a function of depth at various times after the formation of a new semiconductor surface. The example here is a GaAs surface with an electron concentration of $n = 5\times10^{15}\ \mathrm{cm}^{-3}$ and a surface potential of −0.75 V.

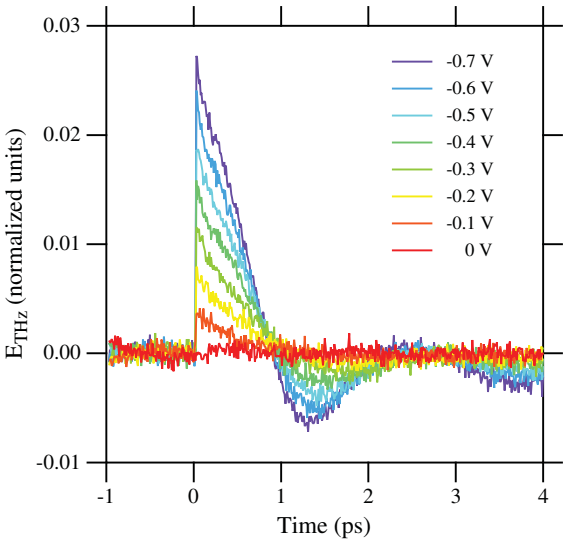

Fig. 2. Electric field of radiation emitted in the far field as a result of the creation of a surface. The material modeled is GaAs ($n = 5 \times 10^{15}$ cm^{-3}). The field is shown for various reasonable values of surface potential.

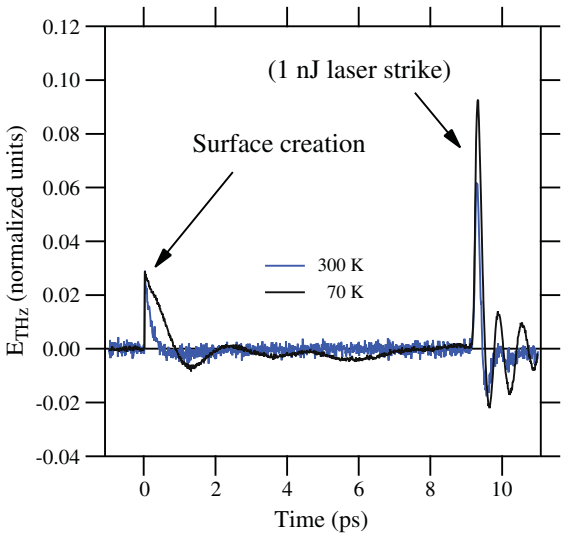

Fig. 4. Electric field of radiation emitted in the far field on creating a GaAs ($n = 5 \times 10^{15}$ cm^{-3} and surface potential of −0.75 V) surface (at time zero) and on optical excitation of the surface with a laser pulse (at elapsed time of 9 ps). It is seen that the magnitudes of the two fields are comparable. Results are given for two temperatures, 300 K and 70 K. At each temperature, the fields arising from surface creation and optical excitation are well above the background noise.

be seen in Fig. 4, the sizes of the electric fields are comparable. Second, we compare the signal with the general noise present. As may be seen from Fig. 4, the signal is substantially greater than the background noise at room temperature (300 K). The signal increases and the noise decreases as the temperature is reduced (70 K), as expected.

We note that the emission of electromagnetic radiation from cleaved semiconductor surfaces has been reported previously [19]. That observation, however, relates to a quite different timescale, μs rather than ps, and a quite different photon energy, eV rather than meV, although the underlying mechanism is bandbending. That work mentioned additional emission on a timescale too fast to be detected with the apparatus used, which may relate to the mechanism proposed here.

In this work we assume that a surface potential will develop within a few fs on the creation of a new surface. We point out that the timescale and exact mechanism (bond rupture/quantum effects) for the formation of the surface potential are not precisely known, but there is no reason why it should not occur on this timescale.

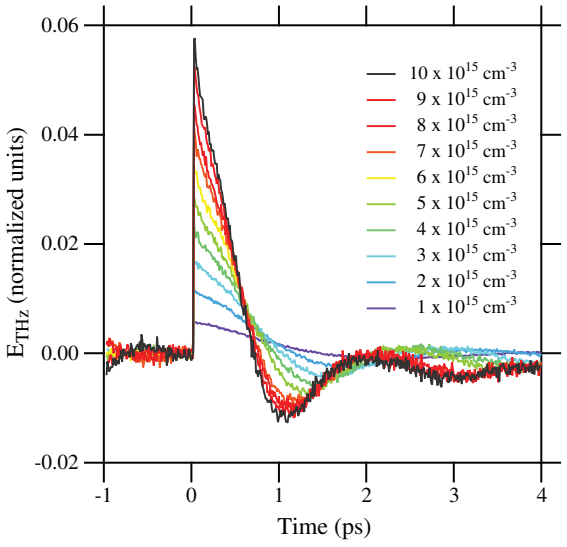

Fig. 3. Electric field of radiation emitted in the far field on creating a GaAs ($V = -0.75$ V) surface. The field is shown for various reasonable doping levels.

In order of magnitude estimations, a surface formed in 1 μs might sustain frequencies to 1 MHz and a surface formed in 1 ns might radiate frequencies to 1 GHz and likewise a 1 ps timescale is needed for 1 THz frequency. We point out that this argument does not continue to shorter timescales and higher frequencies. Our simulations show that even if a surface is created instantly, the charge-carrier dynamics limit the frequency of emission to about 1 THz.

The terahertz emission observed from peeling adhesive tape and attributed to charge separation between the sticky and slippery sides of the tape [6] may in part originate from the mechanism described here. The relative roles of the two mechanisms invite future study through further simulations.

4. Conclusions

We have proposed that charge redistribution on a ps timescale within a newly-created surface may lead to the emission of terahertz-frequency electromagnetic radiation. Increasing either the doping level or the bandbending at the surface enhances the effect. This mechanism is the basis of a new type of terahertz emitter which requires neither optical nor electrical excitation in its operation.

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