Terahertz frequency radiation from Bloch oscillations in GaAs/$Al_{0.3}Ga_{0.7}As$ superlattices

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We have performed a joint theoretical and experimental study to investigate the terahertz radiation from Bloch oscillations in a GaAs/$Al_{0.3}Ga_{0.7}As$ superlattice under the condition that there is no Zener tunneling. The total radiation intensity has been calculated with a semiclassical approach in the low field regime where the Wannier-Stark ladder (WSL) cannot be resolved, and with an exact numerical solution in the high field regime where the WSL is well formed. With an adjustment of the intensity units, without fitting material parameters, the calculated results agree almost perfectly with the measured data given in arbitrary units. Consequently, our work gives convincing evidence that the measured THz radiation is due to the Bloch oscillations.

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The Bloch oscillation (BO) of charge carriers in a periodic potential under an applied uniform electric field $\vec{F}$ was predicted almost 70 years ago.[1,2] In a bulk crystal of lattice constant a, under realistic experimental conditions that the probability of Zener tunneling is negligibly small, the BO amplitude Δ/eF is order of magnitude longer than a, where Δ is the band width. Consequently, in a bulk crystal, before a single BO is completed, the scattering has already destroyed the coherence of the Bloch states. However, for a thin sample of semiconductor superlattice (SL), the SL periodicity d can be very long, the applied F can be very strong, and the miniband width can be very small. Hence, in 1970 Esaki and Tsu[3] proposed to detect BO in such a system.

The two key issues of BO are the structure of the energy levels and the electromagnetic radiation due to the transitions of carriers among these levels. Such a radiation is commonly referred to as BO radiation, which is different from the radiation originated from interband transitions. In the presence of $\vec{F}$, in both the valence band and the conduction band, each miniband decomposes into a Wannier-Stark ladder (WSL) of equal energy separation $h\nu_B = eFd$, where ν_B is the BO frequency.[4] The energy levels of WSL have been resolved in the spectra of optical absorption, photocurrent, and stimulated emission between the valence and the conduction band of GaAs/$Al_{0.3}Ga_{0.7}As$ SL.[5–7] These interband optical detections of the existence of the WSL energy level structure do not prove that the interlevel transition within a WSL has a positive gain. The difficulty of observing the BO radiation was pointed out about ten years ago,[8,9] and we will return to this point later.

The BO frequency ν_B in typical GaAs/$Al_{0.3}Ga_{0.7}As$ SL samples lies in the terahertz (THz) range. A successful detection of BO radiation is then important not only for proving the theoretical prediction of a fundamental physical phenomenon, but also for discovering a new tunable source of coherent THz electromagnetic wave. Using the time-resolved THz emission spectroscopy, THz emission from a GaAs/$Al_{0.3}Ga_{0.7}As$ SL was first observed in 1993.[10] Recently, rapid progress in experiments has been achieved along this direction.[11–14] The existing relevant theoretical works[15–17] are inadequate for a complete interpretation of these experiments, and thus cannot give a theoretical support that these observed THz radiations are indeed BO radiations.

In this Brief Report we will calculate the total intensity $I(F)$ of the THz frequency radiation from the BO in a GaAs/$Al_{0.3}Ga_{0.7}As$ SL under an applied electric field $\vec{F}$, and compare our result with the measured F dependence of the THz intensity. In the high field regime our calculation is exact numerically, since we have checked carefully that in this regime there is negligibly small probability of Zener tunneling. In the low field region our semiclassical approach contains an energy relaxation time τ_e and a momentum relaxation time τ_m, which have been investigated in an independent experiment on the nonequilibrium electron transport in this same sample.[12] The perfect agreement between our calculated $I(F)$ and the experimental data proves that the measured radiation is indeed due to the BO.

The sample which has been investigated experimentally is an undoped GaAs/$Al_{0.3}Ga_{0.7}As$ SL with 73 periods of $d_w = 6.4$ nm thick GaAs wells and $d_b = 0.56$ nm thick $Al_{0.3}Ga_{0.7}As$ barrier layers. The periodicity of the SL is $d = d_w + d_b = 6.96$ nm. Setting the zero reference energy at the GaAs conduction band edge, the barrier height is 250 meV. Under the periodic boundary conditions, the first (lowest) miniband is between 18 meV and 110 meV, while the second miniband is between 154 meV and 439 meV. Hence, only the lower one third of the second miniband lies below the potential barrier. It is well known that under a very strong electric field Zener tunneling through the band gap between two bands may occur,[18] and so complicating the BO phenomena. The Zener tunneling probability can be checked with the well established semiclassical approach Wentzel-Kramers-Brillouin (WKB) approximation.[18] In our theoretical calculation which will be presented later, by checking the degree of interminiband mixing in the eigenfunctions and by using the WKB method to estimate the interminiband tunneling probability, we have concluded that for the electric field strength under consideration, there is no Zener tunneling between the two minibands. Consequently, the second miniband can be ignored.

To model this sample of finite length, we let the growth direction of the sample be the x axis, and nd the positions of the centers of the wells with $n=1,2,\ldots,73$. In the absence of an external electric field, in the Hamiltonian H, the total potential profile consists of two parts $V_{SL}(x)$ and $V_C(x)$. $V_{SL}(x)$ defines the SL potential with $V_{SL}(x)=0$ in the wells, and $V_{SL}(x)=V_0$ in the barriers. $V_C(x)$ is a single square well potential of barrier height V_C and well width $73d$ with $V_C(x)=0$ in the well. This wide well confines our sample to a finite length, and we can imagine that our $GaAs/Al_{0.3}Ga_{0.7}As$ sample of 73 periods is embedded in this wide square well $V_C(x)$. The value of V_C, which will be fixed later, must be much larger than the value of V_0.

We will start with a simple Hamiltonian H_0, which contains only the potential $V_{SL}(x)$. Within the effective mass approximation, using the periodic boundary conditions and taking into account the proper matching conditions at the interfaces, the eigensolutions of H_0 for the conduction band electrons are readily derived. Let $\{b_k(x)\}$ be the corresponding set of Bloch functions in the lowest miniband, which are normalized within one period $0<x\leqslant d$. We will use the basis of Wannier functions $\{a(x-nd)\}$ to solve our problem when the applied electric is strong.[19] The Wannier basis functions are simply

$$a(x-nd)=\frac{d}{2\pi}\int_{-\pi/d}^{\pi/d}dke^{-inkd}b_k(x), \tag{1}$$

where n can be any integer. We choose $b_{-k}(x)=b_k^*(x)$, so the Wannier functions are real.

When an external electric field $\vec{F}$ is applied along the x axis, the field-dependent Hamiltonian is $H(F,x)=H(x)-eFx$. The corresponding eigenfunctions can be expressed as

$$\phi_m(F,x)=\sum_n C_{mn}(F)a(x-nd), \tag{2}$$

where m again can be any integer, although $H(x)$ has only 73 eigenstates. To obtain these 73 eigensolutions, we diagonalize the 73×73 Hamiltonian matrix with field-dependent matrix elements

$$H_{mn}(F)=\int_{\mathrm{SL}}dxa^*(x-md)H(F,x)a(x-nd), \tag{3}$$

where $1\leqslant m,n\leqslant 73$. The above integration runs over the SL sample of 73 periods.

The so-derived eigenenergies, except the two for the surface states, should form a WSL with equal energy separation eFd. It is indeed so when the confinement potential V_C becomes sufficiently large. Furthermore, V_C should be large enough to stop electric field induced ionization. In our calculation, we set $V_C=5$ eV. This value allows us to have convergent eigensolutions for all electric field strength used in the experiment, without field induced ionization. It was pointed out that finite sample size is crucial to a net THz radiation intensity emitted from the WSL.[8,9] However, the two surface states do not contribute to the measured intensity in the THz range because their eigenenergies are largely separated from the levels of the WSL. For the experiment to be considered in the present paper, the energy half width of the pumping laser pulses is approximately 20 meV. Within this energy interval, the levels in the WSL are populated with equal probability. Therefore, in the regime of strong electric field, the total radiation intensity $I(F)$, expressed as $I_h(F)$ with subscript h for high field, is calculated as $I(F)\equiv I_h(F)=AI_{h,0}(F)$ with

$$I_{h,0}(F)=\sum_{(m,n)}(|m-n|edF)\left|\int_{\mathrm{SL}}dx\phi_n^*(F,x)x\phi_m(F,x)\right|^2, \tag{4}$$

where A is a field independent quantity, the value of which depends on the units used. In the above equation, the summation runs over all pairs of WSL eigenfunctions in the electron miniband, excluding the two surface states. Nevertheless, almost all radiation intensity originates from the electron transitions between two adjacent energy levels in the WSL, because the overlap between localized eigenfunctions $\phi_n(F,x)$ and $\phi_m(F,x)$ decreases rapidly as $|m-n|$ increases. Hence, the radiation spectrum is sharply peaked at the BO frequency $\nu_B=eFd/h$.

While the above analysis is for spontaneous emission from the WSL, for which the coefficient A is positive, we must consider scattering effect to study stimulated emission. Let $\epsilon_n(\vec{\rho})$ be the two-dimensional (2D) subband associated with the nth WSL level, and $|n,\vec{\rho}\rangle$ an eigenstate in this subband, where $\vec{\rho}$ is a 2D wave vector. Since the photoexcited electron population on each WSL level is the same, without scattering effect there will be no net stimulated emission from the sample, as was pointed out[8] that the emission due to the vertical transition from state $|n+1,\vec{\rho}\rangle$ to state $|n,\vec{\rho}\rangle$ is perfectly canceled by the absorption due to the vertical transition from state $|n,\vec{\rho}\rangle$ to state $|n+1,\vec{\rho}\rangle$ that occurs at the same frequency and intensity. Scattering will introduce emission process from state $|n+1,\vec{\rho}\rangle$ to state $|n,\vec{\rho'}\rangle$ with emitted photon energy $\epsilon_{n+1}(\vec{\rho})-\epsilon_n(\vec{\rho'})$, as well as absorption process from state $|n,\vec{\rho}\rangle$ to state $|n+1,\vec{\rho'}\rangle$ with absorbed photon energy $\epsilon_{n+1}(\vec{\rho'})-\epsilon_n(\vec{\rho})$. Since the dominating scattering induced processes have $\rho\simeq0$, the emission spectrum will peak at a frequency smaller than ν_B, while the absorption spectrum will peak at a frequency larger than ν_B. Hence, the emitted THz radiation will not be completely reabsorbed within the sample, and a finite intensity will be stimulatively emitted from a WSL. We notice that for both the emission and the absorption process the field-dependent part of the intensity is the same $I_{h,0}(F)$ given by Eq. (4), although different processes have different coefficient values of A. A detailed analysis of this problem including electron-phonon interaction, which will be presented elsewhere, proves that including both emission and reabsorption process, the resultant stimulated emission has a positive coefficient A. Consequently, by using the proper expression of A to include both the spontaneous and the stimulated emission, the total intensity in the high field regime can be written as $I(F)\equiv I_h(F)=AI_{h,0}(F)$ with A positive.

For the case of low electric field such that the WSL separation eFd is less than the level broadening due to scatterings, the electromagnetic wave is emitted when electrons are accelerated in their band motion. The intensity spectrum $I_l(\omega)$, with subscript l for low field, was analyzed[14] to be related to the frequency dependent conductivity $\sigma(\omega)$ as $I_l(\omega)\propto F^2|\sigma(\omega)|^2$. With the semiclassical Boltzmann's equation approach, assuming a tight-binding miniband for the SL, $\sigma(\omega)=\sigma_0\sigma_l(\omega)$ was derived[17] in the form

$$\sigma_l(\omega)=\frac{1}{1+\omega_B^2\tau_m\tau_e}\frac{1-\omega_B^2\tau_m\tau_e-i\omega\tau_e}{(\omega_B^2-\omega^2)\tau_m\tau_e+1-i\omega(\tau_m+\tau_e)}, \tag{5}$$

where σ_0 is a constant. In the above equation, τ_m and τ_e are the momentum and energy relaxation times of electrons, respectively, and $\omega_B=2\pi\nu_B$. Hence, in the regime of low electric field, the total radiation intensity $I(F)$, expressed as $I_l(F)$ with subscript l for low field, is calculated as $I(F)\equiv I_l(F)=BI_{l,0}(F)$ with

$$I_{l,0}(F)=\int d\omega F^2|\sigma_l(\omega)|^2, \tag{6}$$

where B is a field independent quantity, and is positive in semiclsssical theory.

τ_m and τ_e will be estimated from independent experiments. The nonequilibrium electron transport in the same sample which is studied here was investigated in Ref. 12. From the measured transient behavior under various field strength, we deduct that τ_e as a function of the electric field can be approximated as $\tau_e=(1.5–0.025F)$ ps, where F is in units kV/cm. In another experiment[14] on this SL sample, for a given field F lower than 6.5 kV/cm, the measured intensity spectrum $I_l(\omega)$ was fitted with Eq. (6) and Eq. (5), treating τ_m and τ_e as fitting parameters. The so-obtained τ_e is 1.6 ps, and $\tau_m\simeq 0.1\tau_e$. However, we must be aware of the fact that in deriving Eq. (5) the miniband was approximated with a tight-binding band. Therefore, these fitting values $\tau_e=1.6$ ps and $\tau_m\simeq 0.1\tau_e$ provide only a qualitative information. In our calculation we use τ_e around (1.5–0.025 F) ps and the ratio τ_m/τ_e around 0.2, which are reasonable and correct in order of magnitude.

The experimental setup, the measurement method, and the data analysis were given in details in Refs. 12–14. Here we will only outline them briefly. The top contact of the 73 period undoped SL sample was formed by depositing a semitransparent 4-nm-thick NiCr Schottky film, and the bottom ohmic contact was formed by annealing the Au-Ge-Ni alloy. By applying a bias voltage between the top and the bottom electrodes, we can tune an internal electric field F in the undoped SL region.

When a femtosecond laser pulse excites the sample, electron-hole pairs are optically injected into the miniband. Due to an applied electric field F, the carriers start drifting and THz radiation that is proportional to the carrier acceleration is emitted into free space. Since the miniband width for heavy holes is only a few meV, which is much narrower than that for electrons, heavy holes are almost localized. Furthermore, absorption due to light holes is 1/3 of that due to heavy holes. Consequently, electron motion dominates the emitted THz signal.

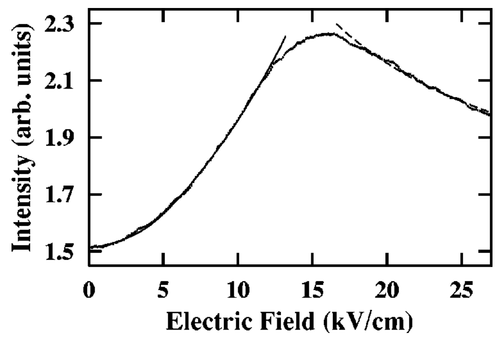

FIG. 1. The radiation intensity as a function of the applied electric field: dotted curve is experimental data, solid curve in low field regime is calculated I(F) using Eq. (6), and dashed curve in high field regime is calculated I(F) using Eq. (4).

Experiments were performed by using 100 fs laser pulses delivered from a mode-locked Al_2O_3:Ti laser. The laser pulses were loosely focused onto the sample surface. The pump photon energy was set to be 1.55 eV, which is close to the bottom of the miniband. With a low pump power of 10 mW, the carrier densities excited in the active region was kept as low as 2×10^{14} cm^{-3} in order to avoid field screening. The generated THz emission was detected by a wideband Si bolometer operated at 4.2 K, whose bandwidth is up to 18 THz. The samples were cooled at $T=10$ K in a continuous flow He cryostat.

In Fig. 1 the dotted curve shows the integrated intensity of the emitted THz radiation as a function of F. In the low field regime the THz intensity rises quadratically. The intensity increases until F reaches about 16 kV/cm, and then starts to roll off. We must point out that the experimental data were presented in *arbitrary units*, which leaves the vertical scale in Fig. 1 undefined. Because of the arbitrary units used in the measured intensity, to compare our calculated $I(F)$ with the measured intensity, we must use the commonly accepted scheme to rescale the units of our calculated results by adjusting the value of A for $I(F)\equiv I_h(F)$, and the value of B for $I(F)\equiv I_l(F)$. The measured intensity also contains a background intensity which depends on the field F very weakly. As indicated in Fig. 1 (in arbitrary units), the background intensity at zero field is 1.51. Hence, we add this constant background intensity to our calculated $I(F)$. Besides such standard method to deal with the undefined units of experimental data, our numerical calculations do not contain any adjustable parameter for material properties. The so-obtained results are shown as the solid curve for the low field region using Eq. (6), and as the dashed curve for the high field region using Eq. (4). In the low field region, the increase of $I(F)$ with F is expected. Since the expression Eq. (6) applies only to semiclassical bands, it naturally begins to break down when the WSL starts to form. By comparing the solid curve and the measured data in Fig. 1, the complete breakdown of the semiclassical theory occurs around $F=12–13$ kV/cm. It is important to point out that for the field strength plotted in Fig. 1, there is no Zener tunneling as we have checked carefully in our calculation. Consequently, in the high field region, with decreasing F, the eigenfunctions of the well defined WSL become more extended and so the adjacent eigenfunctions overlap more. As a result, in this field region $I(F)$ increases with decreasing F. In our exact numerical calculation the broadening of the WSL was not taken into account. Therefore, when the level broadening becomes sufficient as the electric field is reduced, Eq. (4) also breaks down. Again, by comparing the dashed curve and the experiment in Fig. 1, our numerical result is no longer valid around $F=17$–18 kV/cm. To study the crossover region between $F=12$ kV/cm and 18 kV/cm, one needs a better quantum mechanical treatment on scattering induced level broadening.

The excellent agreement shown in Fig. 1 between the experiment (expressed in arbitrary units) and the theoretical results which contains no adjustable material parameter, gives convincing evidence that the measured THz radiation is due to the BO. Such interlevel transitions within a WSL are entirely different from the conventional interband transitions which prove only the existence of WSL level structure. The remaining challenging problem is to derive the correct intensity $I(F)$ for the crossing region from the low field to the high field.

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