Quantum Electronics **37** (12) 1166 – 1168 (2007)

PACS numbers: 41.60. –m; 42.65.Re; 79.60.Jv
DOI: 10.1070/QE2007v037n12ABEH013618

On the mechanism of generation of terahertz electromagnetic radiation upon irradiation of a nanostructured metal surface by femtosecond laser pulses

A.D. Gladun, V.G. Leiman, A.V. Arsenin

***Abstract*.** **A mechanism of generation of coherent terahertz electromagnetic radiation upon irradiation of a nanostructured metal surface by femtosecond laser pulses is proposed. The mechanism is based on the stimulated coherent radiation of electrons emitted from the metal surface and filling hollows in the periodic nanostructure.**

Keywords: *terahertz radiation, femtosecond laser pulse, anomalous photoelectric effect.*

The search for new efficient methods of generating coherent terahertz radiation attracts great recent attention [1 – 6], which is related first of all to various possible practical applications of this radiation [7]. The spectra of many important organic molecules, including proteins and DNA, and phonon resonances of crystal lattices lie in this range. This permits the development of new methods for spectroscopy of biological and semiconductor samples. Terahertz radiation can be used to control chemical reactions and manipulate the electronic states in quantum wells. This radiation is also promising for medical applications for therapy and diagnostics.

At present the most promising direction in the search for methods to generate terahertz radiation is the study of various optical rectification schemes in which petahertz radiation is converted to terahertz radiation [8, 9].

Currently the most popular method for generating terahertz radiation is based on the interaction of femtosecond laser pulses with electrooptical and semiconductor media. Petahertz to terahertz converters based on such media are comparatively low-cost, compact, and can generate radiation powers from a few nanowatts to hundreds microwatts in the spectral range from 0.2 to 2 THz [10]. The relatively low power and low energy conversion coefficient of such sources stimulate the search for more efficient schemes for petahertz – terahertz radiation conversion.

In this paper, we consider a mechanism of generating coherent terahertz radiation, whose nature is determined first of all by the emission of electrons excited by a femtosecond laser pulse from a nanostructured metal surface [4]. This emission can be either thermal emission produced due to nonequilibrium processes excited in a metal by short enough radiation pulses [11] or anomalous photoemission [12 – 14] caused by a considerable enhancement of the field by surface plasmon – polaritons.

The nature of the generation mechanism can be explained by using a simplest model. Consider an infinite periodic structure (lattice) with the period Λ consisting of identical point dipoles with moments $\boldsymbol{p}_n(t)$ arranged along the z axis (Fig. 1). The expression for the polarisation vector $\boldsymbol{P}$ of the structure can be written in the form

$$\boldsymbol{P} = \sum_{n=-\infty}^{\infty} \boldsymbol{p}_n(t)\delta(z - n\Lambda)\delta(y), \quad \frac{\partial}{\partial x} = 0.$$

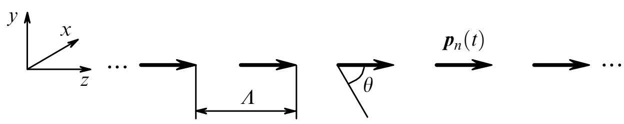

Figure 1. Periodic structure consisting of point dipoles (one-dimensional representation of a lattice with the period Λ).

Consider the radiation of this system at large distances (in the wave zone). The angle θ in Fig. 1 determines the direction to the observation point. Let us introduce the polarisation potential

$$\boldsymbol{\Pi} = \int_V \frac{\boldsymbol{P}(\boldsymbol{r}', t - r/c)}{r} \mathrm{d}\boldsymbol{r}',$$

where $\boldsymbol{r}'$ is the radius vector characterising the dipole position and r is the modulus of the radius vector drawn from the dipole to the observation point. The potentials φ and $\boldsymbol{A}$ of the electromagnetic field are determined by the expressions

$$\varphi = -\operatorname{div}\boldsymbol{\Pi}, \quad \boldsymbol{A} = \frac{1}{c}\frac{\partial \boldsymbol{\Pi}}{\partial t}.$$

The vector $\boldsymbol{\Pi}$ satisfies the wave equation

$$\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\boldsymbol{\Pi} = -4\pi\boldsymbol{P}.$$

For the emitting system under study, we have

A.D. Gladun, V.G. Leiman, A.V. Arsenin Moscow Institute of Physics and Technology (State University), Institutskii per. 9, 141700 Dolgoprudnyi, Moscow region, Russia; e-mail: arsenin@gephys.mipt.ru

Received 5 May 2007; revision received 20 July 2007
Kvantovaya Elektronika **37** (12) 1166 – 1168 (2007)
Translated by M.N. Sapozhnikov

$$\boldsymbol{\Pi} = \sum_{n=-\infty}^{\infty} \frac{1}{r_n} \boldsymbol{p}_n \left(t - \frac{r_n}{c} \right) = \frac{1}{r_n} \sum_{n=-\infty}^{\infty} \boldsymbol{p}_n \left(t - \frac{r_n}{c} \right), \qquad (1)$$

where r_n is the modulus of the radius vector drawn from the nth dipole to the observation point. The latter equality in (1) is justified because $1/r_n$ slowly varies at large distances from the periodic structure. The moment $\boldsymbol{p}_n$ has the components $\{0, 0, p\}$ in the Cartesian system. We assume that

$$p = p_0 \exp[\mathrm{i}(\omega t + k_{\mathrm{g}} \xi)], \qquad (2)$$

where p_0 is the amplitude; k_{g} is the modulus of the wave vector of a wave in the lattice; and $\xi = n\Lambda$ $(n = 0, \pm 1, \pm 2, ...)$. This means that a dipole wave of length $\lambda_{\mathrm{g}} = 2\pi/k_{\mathrm{g}}$ propagates in the lattice. By substituting (2) into (1), we find

$$\Pi = \frac{p_0 \exp[\mathrm{i}\omega(t - r/c)]}{r} \times \sum_{n=-\infty}^{\infty} \exp\left[-\mathrm{i}n\Lambda \left(\frac{\omega}{c} \cos\theta + k_{\mathrm{g}} \right) \right]. \qquad (3)$$

We took into account here that $r_n = r - \xi \cos\theta$.

If the radiation wavelength $\lambda = 2\pi c/\omega \gg \Lambda$, summation in (1) can be replaced by integration, i.e. in this case we can introduce a physically infinitely small interval $\Delta\xi$ satisfying the condition

$$\lambda \gg \Delta\xi \gg \Lambda, \quad \Delta n = \frac{\Delta\xi}{\Lambda},$$

where Δn is the number of dipoles in this interval. By replacing summation in (3) by integration, we obtain

$$\Pi = \frac{p_0 \exp[\mathrm{i}\omega(t - r/c)]}{r} \frac{1}{\Lambda} \times \int_{-\infty}^{\infty} \exp\left[-\mathrm{i}\left(\frac{\omega}{c} \cos\theta + k_{\mathrm{g}} \right) \xi \right] \mathrm{d}\xi$$

$$= \frac{p_0 \exp[\mathrm{i}\omega(t - r/c)]}{r} \frac{2\pi}{\Lambda} \delta\left(\frac{\omega}{c} \cos\theta + k_{\mathrm{g}} \right), \qquad (4)$$

which gives

$$\frac{\omega}{c} \cos\theta = -k_{\mathrm{g}}. \qquad (5)$$

It follows from (5) that $\cos\theta < 0$ and $\omega/k_{\mathrm{g}} = c/\cos\theta$, i.e. the phase velocity of the dipole wave exceeds the speed of light in vacuum.

Consider now emission of a lattice of length l in the wave zone. The emission field of each element of length $\Delta\xi \ll \lambda$ can be represented as a sum of the fields of individual dipoles by neglecting the radiation phase shift. The field of a point dipole in the wave zone is described by the expressions (in the spherical coordinate system $r\varphi\theta$)

$$\Pi = \frac{p_0 \exp[\mathrm{i}\omega(t - r/c)]}{r},$$

$$B_\varphi = \frac{(\mathrm{i}\omega)^2}{c^2} \sin\theta \frac{p(t - r/c)}{r} = \frac{\sin\theta}{c^2 r} \frac{\partial^2}{\partial t^2} p(t - r/c),$$

$$E_\theta = \frac{(\mathrm{i}\omega)^2}{c^2} \sin\theta \frac{p(t - r/c)}{r} = \frac{\sin\theta}{c\, 2r} \frac{\partial^2}{\partial t^2} p(t - r/c), \qquad (6)$$

$$B_r = B_\theta = E_r = E_\varphi = 0,$$

where B_r, B_φ, and B_θ the radial, azimuthal, and polar components of the magnetic induction vector; E_r, E_φ, and E_θ are the radial, azimuthal, and polar components of the electric field vector. According to (6), the total field of a set of Δn dipoles at the length $\Delta\xi = \Lambda\Delta n$ is

$$\mathrm{d}E_\theta = \mathrm{d}B_\varphi = -\frac{\sin\theta}{rc^2} \frac{\mathrm{d}^2 p}{\mathrm{d}t^2} \frac{1}{\Lambda} \mathrm{d}\xi = \omega^2 \frac{\sin\theta}{rc^2} p_0(\xi) \exp(\mathrm{i}\omega t) \frac{1}{\Lambda} \mathrm{d}\xi,$$

where $p_0(\xi)$ is the dipole amplitude in the standing wave field as a function of the coordinate. By integrating over all the elements along the lattice taking the delay $(t \to t - r/c)$ into account, we obtain

$$E_\theta = B_\varphi = \frac{\omega^2}{c^2} \exp(\mathrm{i}\omega t) \int_{-l/2}^{l/2} p_0(\xi) \frac{\sin\theta}{r} \exp\left(-\mathrm{i}\frac{\omega}{c} r \right) \frac{\mathrm{d}\xi}{\Lambda}. \qquad (7)$$

Because the observation is performed far away from the lattice, we can calculate (7) by taking out $\sin\theta/r$ from the integral and make the replacement $r = r_0 - \xi \cos\theta$, where r_0 is the distance from the lattice centre to the observation point. As a result, we obtain

$$E_\theta = B_\varphi = \frac{\omega^2}{c^2} \frac{\exp[\mathrm{i}\omega(t - r_0/c)]}{r_0} \sin\theta \times \int_{-l/2}^{l/2} p_0(\xi) \exp\left(-\mathrm{i}\frac{\omega}{c} \xi \cos\theta \right) \frac{\mathrm{d}\xi}{\Lambda}. \qquad (8)$$

Let us adopt the boundary conditions

$$I(\xi)|_{\xi = \pm l/2} = 0, \qquad (9)$$

where $I(\xi) = \mathrm{d}p/\mathrm{d}t = \mathrm{i}\omega p_0(\xi)$. Condition (9) can be satisfied by assuming that

$$I(\xi) = \begin{cases} \mathrm{i}\omega p_0 \sin\dfrac{\pi s}{l}\xi & \text{for even } s, \\ \mathrm{i}\omega p_0 \cos\dfrac{\pi m}{l}\xi & \text{for odd } m. \end{cases}$$

Integrals in (8) can be readily calculated

$$\int_{-l/2}^{l/2} \sin\frac{\pi s}{l} \xi \exp\left(-\mathrm{i}\frac{\omega}{c} \xi \cos\theta \right) \mathrm{d}\xi = \frac{4\pi s/l}{(\pi s/l)^2 - [(\omega/c)\cos\theta]^2} \sin\left(\frac{\omega}{2c} l \cos\theta \right),$$

$$\int_{-l/2}^{l/2} \cos\frac{\pi m}{l} \xi \exp\left(-\mathrm{i}\frac{\omega}{c} \xi \cos\theta \right) \mathrm{d}\xi =$$

$$= \frac{4\pi m/l}{(\pi m/l)^2 - [(\omega/c)\cos\theta]^2} \cos\left(\frac{\omega}{2c} l \cos\theta\right).$$

Finally, for even s we obtain

$$E_\theta = B_\varphi = \omega^2 p_0 \frac{\exp[\mathrm{i}\omega(t - r_0/c)] \sin\theta}{c^2 r_0 \Lambda}$$

$$\times \frac{\pi s/l}{(\pi s/l)^2 - [(\omega/c)\cos\theta]^2} \sin\left(\frac{\omega}{2c} l \cos\theta\right). \tag{10}$$

For odd m, a similar expression is obtained in which $\sin[(\omega/2c) l \cos\theta]$ is replaced by $\cos[(\omega/2c) l \cos\theta]$.

Thus, we see that, unlike the radiation of a dipole, the radiation fields of a dipole array contain the additional angular dependence, which is determined by the type of dipole oscillations, the lattice length, and the dispersion equation of dipole waves relating ω and $k_g = \pi s/l$.

According to (10), the radiation energy flux density (modulus of the Poynting vector) is

$$S = \frac{\omega^4 p_0^2 \sin^2\theta}{4\pi c^3 r_0^2} \left(\frac{l}{\Lambda}\right)^2$$

$$\times \frac{\sin^2[(kl/2)\cos\theta]}{(\pi s)^2 \{1 - [(k/k_g)\cos\theta]^2\}^2} \cos^2(\omega t - k r_0). \tag{11}$$

Here, $k = 2\pi/\lambda$; $k_g = \pi s/l$; and s is the even number. The energy emitted into the unit solid angle $\mathrm{d}\Omega$ per unit time is

$$\mathrm{d}W = S r_0^2 \mathrm{d}\Omega,$$

where $\mathrm{d}\Omega = 2\pi \sin\theta \mathrm{d}\theta$. If the observation angle satisfies the condition

$$\pm \frac{1}{2} kl \cos\theta = \frac{\pi}{2},$$

i.e. $\cos\theta = \pm\lambda/(2l)$, we obtain for $s = 2$, $\Lambda = 500$ nm, $\lambda = 0.3$ mm ($\nu \approx 1$ THz) and $l = \lambda = 0.3$ mm that $\theta = \pm 60^\circ$ and $l/\Lambda = 600$. This means that the Poynting vector (11) for the lattice radiation exceeds that for the dipole radiation by approximately $(l/\Lambda)^2 = 3.6 \times 10^5$ times.

In conclusion, the natural question arises of how an emitting dipole wave, which is not a surface plasmon–polariton (it is known that the latter does not emit), can be produced on a nanostructured metal surface. This can be performed by an ultrashort laser pulse, which not only excites a plasmon–polariton wave but also causes the emission of electrons oscillating in the wave field. Let us present arguments in favour of this statement.

The optical and kinetic properties of most of the noble metals in a broad temperature range are determined to a great extent by electron–phonon collisions with effective collision frequencies ν_{ep} exceeding electron–electron collision frequencies ν_{ee}. The different situation appears upon the interaction of a high-power femtosecond laser pulse with a metal. After the absorption of a laser pulse, electrons are rapidly heated up to the temperature considerably exceeding the lattice temperature, which remains almost invariable during the pulse action. Already at the electron temperature exceeding two–three thousands kelvins, the condition $\nu_{ee} \gg \nu_{ep}$ is realised. In this case, both the absorption of the laser pulse energy and the heat removal from the skin layer are mainly determined by electron–electron collisions [11, 15].

We can assume that the interaction of a laser pulse with a nanostructured metal surface leads to the strong heating of electrons resulting in their emission. Another reason for electron emission may be the anomalous photoeffect [12–14] due to a manifold enhancement of the field on the metal surface due to excitation of surface plasmon–polaritons. By entering the hollows in the lattice, free electrons oscillate under the action, for example, of the field of the standing long-wavelength plasmon wave excited by the laser pulse in the nanostructure. Electrons oscillating coherently in periodically arranged hollows form the emitting periodic dipole structure.

Thus, we have shown in this paper that electrons emitted by the nanostructured metal surface irradiated by a femtosecond laser pulse play the role of an antenna emitting terahertz plasmons excited by the same pulse. This opens up new possibilities for the interpretation of experimental studies and the development of new pulsed terahertz radiation sources.

Acknowledgements. The authors thank A.P. Kanavin for useful discussions. This work was supported by the Federal Education Agency of the Russian Federation (Grant RNP.2.1.1.6089), Grant MK-4082.2007.2 of the President of the Russian Federation, and by the Russian Foundation for Basic Research (Grant No. 05-01-0790).

References

1. Carr G.L., Martin M.C., McKinney W.R., Jordan K., Neil G.R., Williams G.P. *Nature*, **420**, 153 (2002).
2. Beaurepaire E., Turner G.M., Harrel S.M., Beard M.C., Bigot J.-Y., Schmuttenmaer C.A. *Appl. Phys. Lett.*, **84**, 3465 (2004).
3. Bakunov M.I., Maslov A.V., Bodrov S.B. *Phys. Rev. B*, **72**, 195336 (2005).
4. Welsh G.H., Hunt N.T., Wynne K. *Phys. Rev. Lett.*, **98**, 026803 (2007).
5. Wu H.-C., Sheng Z.-M., Dong Q.-L., Xu H., Zhang J. *Phys. Rev. E*, **75**, 016407 (2007).
6. Kim D.S., Citrin D.S. *J. Appl. Phys.*, **101**, 053105 (2007).
7. Siegel P.H. *IEEE Trans. Microwave Theory Techn.*, **50**, 910 (2002).
8. Auston D.H., Cheung K.P., Valdmanis J.A., Kleinman D.A. *Phys. Rev. Lett.*, **53**, 1555 (1984).
9. Xu L., Zhang X.-C., Auston D.H. *Appl. Phys. Lett.*, **61**, 1784 (1992).
10. Gallerano G.P., Biedron S. *Proc. Conf. on FEL'2004* (Italy, Trieste, 2004) p. 216.
11. Fujimoto J.G., Liu J.M., Ippen E.P., Bloembergen N. *Phys. Rev. Lett.*, **53**, 1837 (1984).
12. Zawadzka J., Jaroszynski D., Carey J.J., Wynne K. *Nucl. Instrum. Meth. Phys. Res. A*, **445**, 324 (2000).
13. Irvine S.E., Dechant A., Elezzabi A.Y. *Phys. Rev. Lett.*, **93**, 184801 (2004).
14. Kupersztych J., Raynaud M. *Phys. Rev. Lett.*, **95**, 147401 (2005).
15. Isakov V.A., Kanavin A.P., Uryupin S.A. *Kvantovaya Elektron.*, **36**, 928 (2006) [*Quantum Electron.*, **36**, 928 (2006)].