Generation of second harmonic of light in crystals with resonant impurities

B. Ya. Zel'dovich and S. D. Kuz'michev
Moscow Physicotechnical Institute

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A light wave induces an oscillating dipole moment in a resonant impurity; its field in the near zone exceeds the macroscopic field and a second harmonic can be efficiently excited on the quadratic nonlinearity of the matrix. A calculation and estimates of the effect are given for the case in which the exciting wave propagates in the self-induced transparency regime.

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In order to generate the second harmonic of light, transparent crystals without a center of symmetry, for which the tensor of quadratic polarizability $\chi^{(2)}$ in the relation

$$P_i^{2\omega}(\mathbf{r})\, e^{-2i\omega t} = \chi_{ikl}^{(2)} E_k^{\omega}(\mathbf{r}) E_l^{\omega}(\mathbf{r})\, e^{-2i\omega t} \tag{1}$$

differs from zero, are usually used. Here the complex amplitudes $\mathbf{A}(r)$ are related to real fields by the relation $\mathbf{A}_{\text{real}}(\mathbf{r},t) = 0.5[\mathbf{A}(\mathbf{r})e^{-i\omega t} + \mathbf{A}^*(\mathbf{r})e^{i\omega t}]$, $\mathbf{P}(\mathbf{r})$ is the dipole moment of the unit volume. Near the absorption bands of the medium the tensor $\chi^{(2)}$ undergoes a resonant increase, but the exciting radiation is strongly absorbed in the medium.

We shall examine a special case in which the medium contains resonant impurity centers. If the amplitude of the induced dipole moment of the impurity is denoted as $\mathbf{d}e^{-i\omega t}$, then at a distance $|r| \lesssim \lambda$ from the center of the field is a sum of macroscopic $\mathbf{E}_{\text{macro}}$ and dipole fields in the near zone:

$$\mathbf{E}(\mathbf{r}) \doteq \mathbf{E}_{\text{macro}} + \frac{3(\mathbf{d}\cdot\mathbf{r})\,\mathbf{r} - r^2\mathbf{d}}{r^5}\,. \tag{2}$$

If we estimate $d \propto \alpha E_{\text{macro}}$, where $\alpha(\text{cm}^3)$ is the polarizability of the center at the resonant frequency, then $|E_{\text{dip}}|/|E_{\text{macro}}| \sim \alpha/r^3$, and for not very large r the dipole field can greatly exceed the macroscopic field, since at resonance we have $\alpha \gg a^3$, where a is an atomic dimension.

We want to point out the fact that this dipole field can efficiently excite polarization (1) at the doubled frequency in a material with a nonresonant matrix. Since the intensity E enters quadratically in (1), the contribution of the dipole field does not disappear when averaging over the volume. As a result, for the case $\mathbf{d} = \mathbf{e}_z d$ and $\chi^{(2)}_{zzz} \neq 0$, for example, we have for the dipole in question the contribution

$$P_z^{2\omega} = \chi^{(2)}_{zzz} \, d^2 \frac{N}{\rho^3}, \tag{3}$$

where N (cm^{-3}) is the density of impurities and the average is performed over the volume of the matrix minus small spheres of radius ρ around the center. As can be seen from (3), the main contribution comes from the immediate vicinity $r \sim \rho$ near the center, and for this reason the cross terms from field of different centers can be ignored.

The large magnitude of the oscillating dipole moment d in (3) is induced by the light wave only in strongly absorbing impurities with a narrow transition line. However, the self-induced transparency effect (SIT, see Refs. 1–3), i.e., passage of radiation along the resonantly absorbing medium without loss of energy, can appear precisely in this kind of impurity centers.

A stationary 2π pulse in the SIT regime has the form[1–3]

$$e^{i(kx - \omega t)} E_{\text{macro}}(x, t) = \frac{2\hbar}{d_{12}\tau} e^{i(kx - \omega t)} \operatorname{sech}[(t - x/u)/\tau], \tag{4}$$

where d_{12} is the matrix element of the dipole transition moment, τ is the duration of the pulse, and $u = u(\tau)$ is the effective velocity. The field (4) satisfies the well-known condition $d_{12}\hbar^{-2} \cdot \int E_{\text{macro}} dt = 2\pi$, while the duration τ must be shorter than all relaxation times.

Assume that the condition for phase synchronization for generation of the second harmonic is satisfied. After substituting the macroscopic and dipole fields into the polarization (1) and (3) and solving the truncated equation for the amplitude $E_2(x,t)$ of the field at the doubled frequency, we obtain

$$E_2(x, t) = i\frac{B\tau}{b}\left\{ \epsilon_1^2 \left[\operatorname{th}\left(\frac{t' + bx}{\tau}\right) - \operatorname{th}\left(\frac{t'}{\tau}\right)\right] - \frac{\epsilon_2^2}{3}\left[\operatorname{th}^3\left(\frac{t' + bx}{\tau}\right) - \operatorname{th}^3\left(\frac{t'}{\tau}\right)\right]\right\}. \tag{5}$$

Here $B = 4\pi(\omega/c)\chi^{(2)}_{zzz}/n$, n is the index of refraction, $\epsilon_1 = 2\hbar/d_{12}\tau$, $\epsilon_2^2 = 3.2\, N d_{12}^2/\rho^3$ $b = v_2^{-1} - u^{-1}$, v_2 is the unperturbed group velocity of the second harmonic, and $t' = t - xv_2 -$ is the local time.

For $bx \gtrsim \tau$ expression (5) describes the increase in the pulse duration of the second harmonic compared to the pumping duration, and $\Delta\tau_2 \propto bx$. This is attributed to the fact that the exciting radiation in the SIT regime moves along the medium, with a lower velocity, while the second harmonic after excitation separates and moves with velocity v_2. Under typical conditions, however, $bx \ll \tau$; then $\tau_2 \sim \tau$ and for the energy coefficient of conversion into the second harmonic we have

$$\eta = \frac{\int |E_2|^2 dt}{\int |E_{\text{macro}}|^2 dt} \approx \frac{4}{3}(Bx\epsilon_1)^2 \left\{ 1 - \frac{2}{15}\left(\frac{\epsilon_2}{\epsilon_1}\right)^2 + \frac{4}{3}\left(\frac{\epsilon_2}{\epsilon_1}\right)^4 \right\}. \tag{6}$$

Here the term $\propto 1$ describes generation of the second harmonic due to the macroscopic field and the term $\propto(\epsilon_2/\epsilon_1)^4$ describes generation due to the dipole field of the centers, while the term $\propto(\epsilon_2/\epsilon_1)^2$ describes their interference. Let us make some numerical estimates. Let $d_{12}\sim 10^{-19}$ CGSE, $\rho\sim 3\times 10^{-8}$ cm, $\tau\sim 10^{-8}$ s, $x=1$ cm, $\omega/c=10^5$ cm^{-1}, and $\chi^{(2)}\sim 10^{-9}$ CGSE; the macroscopic contribution is $\eta_0=4/3(Bx\epsilon_1)^2\approx 10^{-6}$. The contribution of the dipole part becomes appreciable with impurity-center concentration $\gtrsim 10^{14}$ cm^3; thus, for $N=8\times 10^{14}$ cm^3, $\eta\cong 2\eta_0$.

Thus the effects examined above can be observed experimentally. Their study could give information about the properties of resonance impurities and the fields excited by them in the near zone.

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