LASER APPLICATIONS AND OTHER TOPICS IN QUANTUM ELECTRONICS

PACS numbers: 41.75.Ak

DOI: 10.1070/QE2004v034n02ABEH002607

On coherent radiative processes in crystallised ion beams

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Abstract. Periodic spatial structures formed by cooled beams of free ions of the same sign in traps of one or another type are considered. These structures can be treated as quasicrystals. Coherent nuclear radiative processes proceeding in the beams depend on a regular arrangement of nuclei in the structure of such crystallised ion beams. These processes include the Dicke superradiance, directional emission of gamma quanta, the establishment of a distributed feedback, and the appearance of the hidden inversion of nuclear states. Conditions required for the emergence of these effects are quantitatively estimated.

Keywords: crystallised ion beams, coherent radiative processes.

A regular behaviour of beams of free charged particles of the same sign (electrons, ions, etc.), allowing one to treat them like regular crystals [1], is determined by a few basic counteracting factors. These are the Coulomb repulsion, thermal motion and the influx of new particles into the beam, which specifies their averaged concentration. Of course, such quasi-crystals are not stable stationary structures, and their maximum lifetime is determined either by the time during which they are kept in a trap or by the duration of propagation of a given group of particles from their source to a catcher. However, when new particles are continuously supplied into the beam, thereby maintaining the structure of a moving quasi-crystal lattice, the latter can be treated as a conditionally stationary lattice. Therefore, a regular periodic arrangement of particles in the beam should be manifested in a variety of radiative processes proceeding in them, in particular, in gamma transitions in nuclei of ions in the beam [2]. Interest in these problems and the study of conditions to which the beams should correspond was revived due to successful experiments on the formation and observation of such cooled crystal structures both in high-energy ion beams (in particular, in storage rings) and relatively low-energy beams [3-18]. The known advances in laser manipulations (cooling) with

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Received 29 October 2003 *Kvantovaya Elektronika* **34** (2) 180 – 184 (2004) Translated by M.N. Sapozhnikov neutral atoms and ions [19] also stimulate the investigation of these problems.

By now one- and three-dimensional crystal ion structures have been observed. However, as the simplest model of a quasi-crystal beam of charged particles [1], it is convenient to consider an extended one-dimensional chain with the average interval \bar{a} between particles and the diameter of the beam (doubled deviation of individual particles from the beam axis) $d \ll \bar{a}$, which is confined by a transverse potential well of one or other type (for example, a longitudinal magnetic field).

The average interval \bar{a} is determined by the beam current J, the ion multiplicity q, and the average transfer velocity of ions v:

$$\bar{a} = \frac{eqv}{J} \tag{1}$$

where e = |e| is the elementary charge. For example, $\bar{a} = 10^{-2}$ cm for q = 1, $v = 10^{7}$ cm s⁻¹, and J = 0.16 nA (below, all numerical estimates use these parameters, unless otherwise stated).

When positive ions are uniformly arranged on the beam axis and spaced by the interval \bar{a} , the electric potential of the chain in the vicinity of the point z=0, $\rho=0$ of location of one of the ions (after the subtraction of its own potential) is saddle-shaped in the cylindrical coordinate system and is described for small deviations from z=0 and $\rho=0$ (where the potential is normalised to zero) by the expression [1]

$$U(z,\rho) \approx -0.3 \frac{eq}{\pi \varepsilon_0 \bar{a}} \left\{ \left(\frac{\rho}{\bar{a}} \right)^2 \left[1 - 0.65 \left(\frac{\rho}{\bar{a}} \right)^2 + 5.2 \left(\frac{z}{\bar{a}} \right)^2 \right] - 2 \left(\frac{z}{\bar{a}} \right)^2 \left[1 + 0.85 \left(\frac{z}{\bar{a}} \right)^2 \right] \right\}, \tag{2}$$

where ε_0 is the permittivity of a vacuum.

The saddle-shaped potential $U(z, \rho)$ has at the point z = 0, $\rho = 0$ the minimum over the coordinate z and the maximum over the coordinate ρ . Therefore, as pointed out above, the transverse instability of the beam should be suppressed with the help of a trap of one or other type, for example, by applying a longitudinal magnetic field. In the latter case, the confining value of the magnetic field can be estimated from the balance of radial forces (Coulomb, Lorentzian, and centrifugal):

$$H \approx \frac{2}{\bar{a}^{3/2}} \left(\frac{0.6Mc^2}{\pi\mu_0}\right)^{1/2},$$
 (3)

where M is the ion mass; c is the speed of light; and μ_0 is the permeability of a vacuum. For an ion with the isotopic number A = 100, the magnetic field strength H is 1.26 kOe.

The absolute longitudinal ordering of the beam cannot be established due to the thermal motion of particles (even when the equilibrium state is established), which results in the average deviation from the equilibrium longitudinal coordinate z=0 by the value

$$|z| \approx \frac{\bar{a}}{eq} \left(\frac{\pi \varepsilon_0 \bar{a} k T}{1.2}\right)^{1/2},$$
 (4)

where T is the absolute 'longitudinal temperature' of the beam and k is the Boltzmann constant. We can assume that the regular arrangement of ions in the chain, which means the formation of a crystal, is established, if $|z| \leqslant \bar{a}$, i.e., when the beam is sufficiently cooled and

$$(kT)^{1/2} \ll eq \left(\frac{1.2}{\pi \varepsilon_0 \bar{a}}\right)^{1/2}.$$
 (5)

This gives the estimate $T^{1/2} \ll 1K^{1/2}$.

Ions execute longitudinal oscillations in the potential well (2) with respect to the equilibrium position (z = 0) with the frequency

$$\Omega = eq \left(\frac{1.2}{\pi \varepsilon_0 M \bar{a}^3}\right)^{1/2}.$$
 (6)

In addition, when the transverse magnetic confinement is used, one more resonance is present at the cyclotron frequency

$$\Omega_{\rm c} = eq \left(\frac{2.4}{\pi \varepsilon_0 M \bar{a}^3} \right)^{1/2} = \sqrt{2} \Omega. \tag{7}$$

This gives the estimates $\Omega \approx 80$ kHz and $\Omega_{\rm c} \approx 110$ kHz.

Both types of low-frequency oscillations are strictly speaking decaying because ions emit electromagnetic waves at the corresponding frequencies. Thus, if weak longitudinal oscillations are treated as purely dipole oscillations, then the decay time

$$\tau = \frac{5(\pi c)^2 \varepsilon_0}{(\mu_0/\varepsilon_0)^{1/2}} \frac{M^2 \bar{a}^3}{(eq)^4} \tag{8}$$

proves to be quite large both compared to the oscillation period $(\tau \gg 2\pi/\Omega)$ and other characteristic times of the effects analysed below. This allows us to assume that oscillations almost do not decay. Therefore, a weak radiative cooling of the ion chain due to a decrease in the oscillation energy of ions can be noticeable only after a long time, for example, in a storage ring with a sufficiently long storage time. The lower limit of such cooling is determined by the ambient temperature, which can be reduced by placing the ion chain into a cryostat.

By the way, because the radiation pattern of a dipole is determined by the dependence $\sim \sin^2 \theta$ (where θ is the angle measured from the ion-chain axis), the radiation of ions at the frequency Ω along the chain, which could perturb the motion of neighbouring ions, is absent.

The above oscillation processes can complicate the spectrum of radiative transitions between the levels of ions and nuclei (in particular, the Doppler splitting of

the lines) only if the values of Ω and (or) Ω_c are comparable with the natural widths of the transitions and their inhomogeneous broadening is strongly suppressed. The latter condition can be achieved by the laser cooling of an ion beam (see, for example, Ref. [19]) or by using specific electro- kinematic methods for producing monokinetic ion and atomic beams [20, 21].

The periodicity of quasi-crystal ion beams can be revealed in radiative processes proceeding in them under conditions that are determined by the type of these processes.

The Dicke superradiance can arise when a set of emitters is located within a spatial region with dimensions noticeable smaller than the wavelength (see, for example, Ref. [22]). It is obvious that this spatial restriction prevents the direct observation of superradiance in the gamma range, where the wavelength is comparable to the atom size or can be even much smaller. However, it is known that a regular arrangement of a set of emitters with a period that is equal or a multiple of the wavelength eliminates this obstacle, by 'combining' remote emitters at the same point in the wave phase. In this case, the requirement of accuracy of the in-phase location of emitters imposes a new (along with the condition $|z| \ll \bar{a}$) restriction on the deviation of ions from their equilibrium position (z = 0) in the chain compared to the emission wavelength λ ($|z| \le \lambda$) and, hence, on the beam temperature,

$$(kT)^{1/2} \ll eq \left(\frac{1.2}{\pi \varepsilon_0 \bar{a}}\right)^{1/2} \frac{\lambda}{\bar{a}}.$$
 (9)

This restriction differs from (5) by a small factor λ/\bar{a} . Thus, for $\lambda=10^{-7}$ cm, deep cooling providing the inequality $T^{1/2} \ll 10^{-5} \ \mathrm{K}^{1/2}$ is required.

The increased directivity of coherent radiation arises when the same temperature restriction is fulfilled for a regular set of emitters, which can be treated as an in-phase sequence of dipoles oscillating perpendicular to the chain axis and having the radiation pattern of the type $\sim \cos^{2N} \vartheta$, where N is the number of ions on a smaller of two linear scales (the coherence length or the chain length) and ϑ is the angel measured from the chain axis.

The distributed feedback between emitters can exist upon stimulated emission (provided the same temperature and spatial restrictions are fulfilled) due to regular scattering of hard coherent radiation from the electron shells of periodically arranged atoms.

The hidden nuclear inversion (see, for example, Ref. [23]) can be caused, as is known, by the mutual shift of gamma absorption and emission lines, whose energy proves to be higher or lower, respectively, than the nuclear transition energy E_0 by the recoil energy of the nucleus

$$E_{\rm rec} = \frac{E_0^2}{2Mc^2} \tag{10}$$

or by the kinematic (depending on the observation direction) Doppler shift due to the additional recoil velocity

$$|\Delta v| = \frac{E_0}{Mc} = 2c \frac{E_{\text{rec}}}{E_0} \tag{11}$$

acquired by the nucleus, which is collinear to the wave vector of a gamma quantum.

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The structure of an ion crystal is preserved if the recoil energy $E_{\rm rec}$ (10) is small compared to the depth of the potential well (2), i.e.,

$$\frac{E_0^2}{2Mc^2} < eqU(z, \rho = 0) = 0.6 \frac{(eq)^2}{\pi \varepsilon_0 \bar{a}} \left(\frac{z}{\bar{a}}\right)^2, \tag{12}$$

where $|z|/\bar{a} \ll 1$. This imposes the restriction

$$E_0 < eq \frac{|z|}{\bar{a}} \left(\frac{1.2Mc^2}{\pi \varepsilon_0 \bar{a}} \right)^{1/2} \tag{13}$$

on the gamma transition energy. Thus, $E_0 < 8|z|/\bar{a}$ for A=100 (where \bar{a} is in centimetres, and E_0 is in kiloelectronvolts.)

Because ions are rather weakly bounded in a crystal chain, the energy splitting between the gamma absorption and emission lines by the doubled recoil energy $2E_{\rm rec}$ is manifested as in free nuclei. At the same time, the kinematic component of the shift of gamma lines acquires new features in a crystal chain of ions containing nuclei, because, strictly speaking, the recoil momentum transfers to the oscillational degree of freedom of the ion rather than to its translational movement, as in the case of free nuclei, which is usually studied [23]. Here, several particular cases should be analysed.

If the lifetime of a nuclear transition is substantially shorter than the oscillation period of ions in a crystal,

$$\tau_{\gamma} \ll 2\pi/\Omega,$$
 (14)

so that the nuclear radiative process 'has no time' to response to nuclear oscillations, the situation is close to the case of free nuclei, and the kinematic Doppler shift is specified by velocity Δv (11).

When the lifetime τ_{γ} of a gamma transition and the oscillation period $2\pi/\Omega$ of ions are comparable, the harmonic nature of the nuclear motion is still not revealed in full measure because it does not contain a sufficient number of periods $2\pi/\Omega$. However, the instant of observation of the radiative transition line proves to be essential. Indeed, the nucleus acquires the maximum recoil velocity (11) at the instant of the initial radiative transition that gives rise to this velocity. If, for example, the acquired velocity $|\Delta v|$ (11) is directed parallel to the ion-chain axis, the ion containing the nucleus slows down in the potential field of the chain from the velocity $|\Delta v|$ down to full stop at the end of the first quarter of the oscillation period of ions.

Then, the ion velocity increases up to its maximum value $|\Delta v|$, etc. The value and sign of the kinematic Doppler shift of the lines 'following' a change in the ion velocity will change correspondingly. The picture becomes even more complicated when the recoil velocity (11) is not parallel to the chain axis.

Finally, when inequality (14) has the opposite sign,

$$\tau_{v} \gg 2\pi/\Omega$$
 (15)

(in a simple case of the collinear wave vectors of absorbed and emitted gamma quanta to the chain axis, which takes place upon excitation of nuclei by well-collimated longitudinal beam of incoherent X-rays and stimulated emission of gamma quanta along the chain axis), ion oscillations excited by the recoil momentum of the gamma quantum

produce the low-frequency Doppler modulation described by the expression

$$\omega = \frac{E_0 \pm E_{\text{rec}}}{\hbar} \left(1 + \frac{|\Delta v|}{c} \cos \Omega t \right)$$

$$\approx \frac{E_0}{\hbar} \left(1 \pm \frac{E_{\text{rec}}}{E_0} + \frac{|\Delta v|}{c} \cos \Omega t \right). \tag{16}$$

As a result, a discrete gamma spectrum appears with frequencies

$$\omega_n = \omega_0 + n\Omega \quad (n = 0, \pm 1, \pm 2...)$$
 (17)

and the central frequency

$$\omega_0 = \frac{E_0 \pm E_{\text{rec}}}{\hbar},\tag{18}$$

which coincides with the lines shifted by the recoil energy (10). The upper and lower signs in (18) and in the first factor in (16) correspond to the absorption and emission gamma lines, respectively. Of course, as mentioned above, the lines in the spectrum (17) can be resolved only if the inhomogeneous thermal broadening is suppressed to the value that is much lower than the interval between the lines. This imposes one more restriction on the ion-beam temperature:

$$(kT)^{1/2} \ll \frac{c\hbar eq}{2E_0} \left(\frac{0.6}{\pi \varepsilon_0 \ln 2} \frac{1}{\bar{a}^3}\right)^{1/2}.$$
 (19)

For A = 100 and $\lambda = 10^{-7}$ cm ($E_0 = 1.24$ keV), we obtain $T^{1/2} \ll 10^{-6}$ K^{1/2}, which proves to be a more strict restriction than (9).

The presence of some or other lines in the spectrum (17) is determined by the order of the sequence of radiative transitions in nuclei of the ion chain. If all the nuclei are not excited initially, the Doppler frequency modulation (16) is absent, and all the terms with |n| > 0 are absent in the absorption spectrum [17]. These terms appear only in the emission spectrum of excited nuclei due to excitation of ion oscillations upon absorption of a gamma quantum (the latter can be treated as the pumping of nuclei by X-rays in the two-level scheme [23]). Then, the hidden nuclear inversion of states appears if none of the emission lines coincides with the absorption spectrum, i.e., if

$$E_{\rm rec} \neq \frac{|n|}{2}\hbar\Omega\tag{20}$$

or if

$$\frac{E_0^2}{c^2\hbar e a} \left(\frac{\pi \varepsilon_0 \bar{a}^3}{1.2M}\right)^{1/2} \neq |n|. \tag{21}$$

This condition is readily satisfied because the left-hand side of (21) is so large that it can coincide with an integer |n| only when $|n| \ge 1$, i.e., for a very remote line whose amplitude is close to zero. Therefore, the hidden inversion is established for almost all emission lines with moderate values of |n|.

However, the presence of many emission lines, which do not coincide with the resonance absorption lines of the unexcited nuclei, is hardly a positive factor because, for a finite total number of excited nuclei, this leads to a relative decrease in the gain in each of the lines. Therefore, some useful rarefaction of the line spectrum proves to be possible, taking into account that the amplitudes of lines in the emission spectrum are proportional to quadratic terms of a series of the Bessel functions $J_n^2(\mu)$ of the first kind with frequencies (17) and arguments

$$\mu = \frac{|\Delta v|}{c} \frac{\omega_0}{Q} \tag{22}$$

playing the role of the modulation index [24].

The modulation index (22) is an essentially positive quantity $(\mu > 0)$. Therefore, the amplitude J_0^2 of the main line in the spectrum with n = 0 is always smaller than unity, but it can be set quite close to unity assuming that the modulation index $\mu \ll 1$, i.e., if

$$\left(\frac{\lambda}{\bar{a}}\right)^{3/2} \geqslant \frac{\pi^2}{eq} (6.7c\varepsilon_0 \hbar)^{1/2} \left(1 - \frac{E_{\text{rec}}}{E_0}\right) \times \left(\frac{E_0}{Mc^2}\right)^{1/2} \approx \frac{210}{q} \left(\frac{E_0}{Mc^2}\right)^{1/2}.$$
(23)

Thus, $(\lambda/\bar{a})^{3/2} \gg 0.02$ for A = 100 and $E_0 = 1$ keV. At the same time, for $\mu \ll 1$, the amplitudes of all other spectral lines with |n| > 0 prove to be much smaller than unity.

However, the modulation index μ can be also large even when $|\Delta v|/c \ll 1$, because $\omega_0/\Omega \gg 1$. This leads to an important result [24]: since the Bessel series $J_n^2(\mu)$ of the quadratic amplitudes of the lines with frequencies ω_n (17) and the specified modulation index changes nonmonotonically with the index n, the lines with zero amplitudes

$$J_n(\mu_r) = 0 (24)$$

can appear in it, if the argument of the Bessel function J_n of the first kind of the order of n coincides with some of its roots μ_r :

$$\mu = \mu_n = \mu_r. \tag{25}$$

This allows one to remove undesirable lines from the 5 spectrum.

For example, the line with |n| = 1 will be completely suppressed for $\mu_1 = 3.83$ (the first root), while the quadratic amplitude of the next term with |n| = 2 will amount to $\sim 15\%$ of the main line. Of course, other combinations, which can be more optimal, are also admissible.

The requirement (25) imposes specific conditions on the quantities entering expression (22) for the modulation index:

$$\frac{\bar{a}}{\lambda} = \left[\frac{0.3 (eq\mu_r)^2}{2\pi^4 c \varepsilon_0 \hbar (1 - E_{\text{rec}}/E_0)^2} \frac{Mc^2}{E_0} \right]^{1/3}$$
16.

$$\approx 0.03 \left(\frac{Mc^2}{E_0} q^2 \mu_r^2\right)^{1/3}.$$
 (26)

Thus, for A = 100, $E_0 = 1$ keV, and $\mu_r = 3.83$, we obtain $\lambda/\bar{a} \approx 0.03$.

Upon anti-Stokes optical excitation of isomer nuclei from a long-lived metastable state to a higher-lying additional level followed by emission of gamma quanta to the ground state (see, for example, Ref. [23]), the general properties of processes under study do not change compared to the 'two-level scheme' considered above. The exception is that the transition energy E_0 in expression (10) for the recoil energy should be replaced by the energy of a triggering transition from the metastable state to the upper additional level, which should be taken into account in all the following expressions containing $E_{\rm rec}$.

In this connection, it is pertinent to mention experimental attempts that are being continued to observe the anti-Stokes transitions in metastable nuclei triggered by an incoherent X-ray flux using both radiation from X-ray tubes and relativistic electrons (see Refs [25–28] and other papers). There is reason to believe that the extension of these experimental attempts (the choice of other isomer nuclei and the use of other triggering sources) can appreciably improve the reliability of the results obtained.

For example, undoubtedly of interest is the isomer nucleus $^{242}_{95}\mathrm{Am^m}$ with the metastable-state energy of 48.63 keV, the lifetime of 121 year, and the energy of the triggering transition to the higher-lying additional level as low as 4.27 keV. Note that the triggering energy for this nucleus and the energy of one of the electronic transitions in its ion shell coincide, and this resonance can enhance the triggering transition.

A low value of the triggering energy for the nucleus $^{242}_{95}\mathrm{Am^m}$ (and possible for some other isomer nuclei) and successful experiments on excitation of low-lying nuclear levels in a dense hot plasma formed over a solid target in the focus of a high-power pulsed laser beam (see Ref. [29] and other papers) stimulate the experimental study of possible anti-Stokes nuclear transitions using a laser plasma as an alternative triggering source.

Acknowledgements. This work was partially supported by the ISTC (Grant No. 2651p) and the US CRDF-RF Ministry of Education Award VZ-010-0.

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