New kinetic technique for studying coherent optical effects

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The main problem in currently projected studies of coherent time-dependent effects is related to the attempt to move into the short-wavelength part of the optical spectrum. It is already quite clear that conventional schemes, media, and techniques for designing an experiment are inadequate to solve this problem. In the present work a new concept is studied theoretically, based on the idea of coherent excitation of a beam of ions by spatially periodic structures.

1. INTRODUCTION

At present, efforts to investigate coherent electromagnetic processes are concentrated in the search for new techniques for coherent excitation, new media, and methods of observing coherent optical effects. The growth of efforts in this direction may yield new results in the physics of cooperative processes, and extend the possibilities of obtaining spectroscopic and other physical information in this area of investigation.

There are several techniques for exciting coherent optical signals, depending on the scientific and practical problems and also on the type of resonant medium. Historically, the first and standard techniques consists of applying pulsed laser light.[1] Another method was first employed by Buwer and Shoemaker.[2] In this variant the exciting laser operates in the cw mode, and light absorption by a specific subsystem of dipole particles "turns on" or "turns off" due to the Stark effect. The method proposed by Vlasov *et al.*[3] is based on a fast sweep over the sample with a continuous laser beam. The so-called kinetic technique for exciting coherent optical effects is applicable to an ensemble of moving particles.[4] This technique was used mainly to treat atomic and molecular beams excited by laser light. Manykin and Gorshokov[5] outlined a number of new ideas which allow the range of techniques for optical excitation of coherent collective states in resonant media to be extended.

In this work we present theoretical analysis of these and other techniques for kinetic studies of coherent optical effects.

2. RESONANT ION BEAM

Consider a beam of accelerated ions in a resonant medium. The basic elements of the accelerating system are the ion source and channels for shaping, transporting, and moving the beam to the target. Present-day accelerators allow particles to be accelerated to relativistic energies, confine the angular divergence to the smallest possible values, and stably regulate the magnitude and current density so as to obtain ion beams of practically every element of the periodic table over a range of charged states.

As a collection of directed charged particles an ion beam is characterized by the following dynamical parameters: the type and charge of ions; the pulse length (for an ion pulse, the number of ions in the pulse, the duration of the pulse, and the space–time form; for a continuous beam, the current, charge density, and size and shape of the cross section); the energy E; the relative energy spread $\Delta E/E$; and the collimation angle θ.

As a resonant medium an ion beam is characterized by the following optical spectroscopic properties: the number of resonant optical centers N; the size, shape, and nature of the distribution of resonant centers; two separate states for each center, usually with a dipole transition moment d_{mn} and transition frequency ω_{mn}; a spectral absorption (emission) line for the resonant system with inhomogeneous broadening $\Delta\omega_n=2\pi/T_2^*$, homogeneous broadening $\Delta\omega_{\text{homo}}=2\pi/T_2$, and natural width $\Delta\omega_{mn}=2\pi/T_1$, where T_2^*, T_2, and T_1 are the reversible phase, irreversible phase, and energy relaxation times respectively.

In beam theory real streams of particles can be represented as diverging, converging, and parallel laminar ion beams. Depending on the shape of the transverse cross section we distinguish sheet, axisymmetric, hollow, and other beams.

In real ion emission processes the particles leave the source with velocities that differ in magnitude and direction. The inhomogeneous broadening may be produced by an inhomogeneous external field in the ion-optical system of the accelerator also. In the first case the distribution of resonant particles can be described by a Gaussian, $g(\Delta\omega)=\exp(-(\Delta\omega^2)/\Delta^2)/\Delta\sqrt{\pi}$, where $\Delta\sqrt{\pi}$ is the characteristic half-width of the distribution and we have written $\Delta\omega=\omega-\omega_0$ (ω_0 is a characteristic frequency of the spectrum). In the other case, the corresponding resonant particle distribution function is given by a Lorentzian line shape $g(\Delta\omega)=\Delta/\pi(\Delta^2+\omega^2)$. Estimates show that

$$T_1\sim 10^{-6}\text{–}10^{-11}\ \text{s},\quad T_2^*\sim 10^{-9}\text{–}10^{-13}\ \text{s}.$$

In a beam the ion energy spectrum agrees very closely with the energy levels of the individual particles. This fact enables us to predict ahead of time many of the possible resonant transition schemes in the medium. Moreover, the parameters of the resonant ion beam can easily be varied even in the course of a single experiment. Hence the ion

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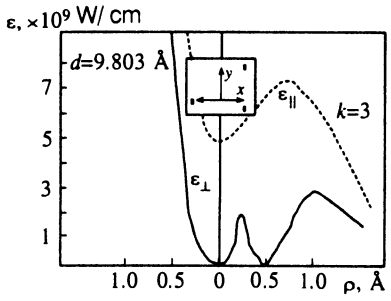

FIG. 1. Electric field components in the Møller approximation for the atomic potential for the ⟨111⟩ channel in a Ge crystal as a function of the distance ρ from the center of the channel; k is the index of the Fourier harmonic.

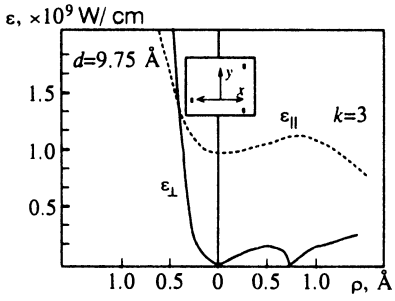

FIG. 2. Electric field components in the Møller approximation for the atomic potential for the ⟨111⟩ channel in a NaCl crystal as a function of the distance from the center of the channel; k is the index of the Fourier harmonic.

beams are close to the ideal situation, which enables us to obtain resonant media with random, uniform, or ordered distributions of optical centers.

3. SPATIALLY PERIODIC ELECTROMAGNETIC FIELDS

Many techniques for creating quantum coherence are known. One way or another, they all involve the use of some kind of external perturbation. For an ion beam this perturbation may involve spatially periodic electromagnetic fields, e.g., a crystal or the surface of a crystal.

A crystal consists of a three-dimensional periodic structure containing a large number of atoms, which in turn produce strong Coulomb fields partly shielded by the electron subsystem (Fig. 1). The field inside a crystal oscillates continuously with a small amplitude about its mean value due to oscillations of the crystal lattice ($U_\perp \sim 0.1$–0.01 Å). The amplitude of the crystal lattice oscillations increases with temperature, but is always much smaller than the lattice constant ($d \sim 1$–10 Å). The symmetry of the field inside the crystal is determined by the crystal symmetry; it depends on the deformation, defects, and polarization of the crystal.

We will use the term "surface" of the crystal to refer to the monatomic layer which separates the crystal from the outside world. A real surface has some degree of impurities due to adsorption of particles from the surrounding environment and their diffusion into the interior of the material. However, in an ultrahard vacuum ($\geqslant 10^{-8}$ Pa) it is possible to obtain and preserve clean crystal surfaces for a relatively long time ($\geqslant 10^4$ s).[8] The crystallography of a clean surface is more varied than that of the interior, but it is also characterized in terms of uniformity and order. The strength of the periodic atomic fields at the surface of a crystal almost reaches the values of the fields inside the crystal (Fig. 2).

Another example of a spatially periodic electromagnetic field is that of a laser wave, a source of coherent optical emission. Present-day laser technology allows continuous and pulsed electromagnetic waves in the visible and nonvisible wave bands to be obtained with field strengths that sometimes reach those of interatomic fields ($E_0 \sim 10^8$ V/cm). The laser radiation is "quasimonotonic" ($\Delta\omega/\omega \sim 10^{-5}$–$10^{-7}$), the wave front has a large radius of curvature (divergence $\sim 10^{-4}$), and the field amplitude changes abruptly on the wave front (it has the shape of a Gaussian). In the general case the shape of a laser pulse can be arbitrary. In calculations, however, it is often sufficient to use a harmonic plane wave and to replace a complicated pulse form with a square wave or triangular profile, or to use pulses with hyperbolic or Gaussian envelopes.

As an example of a generalized system of spatially periodic fields such as those described above we can consider a wiggler. A wiggler is a device in which spatially periodic electromagnetic fields are produced. Examples include a static electromagnetic field with alternating directions, a high-power laser field, an intense standing wave, the field inside a crystal, etc. A transverse harmonic electromagnetic field of length $l=Nd$, where N is a number and d is the length of a period of the wiggler, can serve as a mathematical model of a wiggler, as can the field of a plane linearly polarized wave with spatial period of length $d=c/\omega_0$.

4. CONDITIONS FOR COHERENT STATES

Coherent optical effects arise as the result of the interaction of the resonant medium with electromagnetic waves under certain conditions. Consider the interaction between an ion beam and a wiggler. We look for the conditions for resonant coherent excitation of the collection of ions. Note that the interaction between the ions and wigglers of various types has quite specific properties. Consider, e.g., the interaction between ions and the crystal surface in the course of specular reflection.

Let θ be the grazing-incidence angle of an ion parallel to an atomic chain on the surface of the crystal at an angle less than the critical angle $\hbar_c$ for axial channeling:

$$\theta \leqslant \psi_c = (2z_1 z_2 e^2/dE)^{1/2}. \quad (1)$$

Here $z_1 e$ and $z_2 e$ are the ion charge and the atomic charge respectively and E is the particle energy. In this case the overwhelming majority of the ions undergo specular reflection from the surface.[9]

For the energy of the interaction of the ion with the periodic field of the atoms we can write

$$U(b,x)=U_0(b)+\sum_{k=1}^{\infty}U_k(b)\cos k\Omega x, \tag{2}$$

where

$$U_0(b)=\frac{1}{d}\int_{-\infty}^{\infty}V[(b^2+x^2)^{1/2}]dx,$$

$$U_k(b)=\frac{2}{d}\int_{-\infty}^{\infty}V[(b^2+x^2)^{1/2}]\cos k\Omega x\,dx,$$

$V[(b^2+x^2)^{1/2}]$ is the ion–atom interaction energy, the x axis is directed parallel to the chain, and we have written $\Omega=2\pi/d$.

At relatively small distances b between the ion and one of the chains the effect of a neighboring chain can be neglected and we can assume that the ion moves in the field of a single chain of atoms. In the Møller interaction potential the constant component and the amplitudes of the harmonics are equal:

$$U_0(b)=\frac{2z_1z_2e^2}{d}\sum_{i=1}^{3}\alpha_iK_0\left(\frac{b\beta_i}{a}\right),$$

$$U_k(b)=4z_1z_2e^2[(1+\delta k\Delta)d]^{-1}\sum_{i=1}^{3}\alpha_iK_0\left\{b\left[\left(\frac{b}{a}\right)^2+\left(\frac{2\pi k}{d}\right)^2\right]\right\}. \tag{3}$$

Here $a=0.8853a_0(z_1^{-2/3}+z_2^{-2/3})^{-1/2}$ is the Coulomb shielding radius, the constants are $\alpha_i=\{0.1;0.55;0.35\}$, $\beta_i=\{6;1.2;0.3\}$, $K_0(...)$ is the modified Bessel function of order zero, and $k=1, 2, 3, \ldots$.

The ion motion parallel to the atomic chain can be assumed to be approximately uniform with velocity v. Then we have $x=vt$ and the sum in Eq. (3) can be treated as a periodic perturbation in time:

$$U(b,t)=\sum_{k=1}^{\infty}U_k(b)\cos\omega_kt, \tag{4}$$

where the frequency is $\omega_k=2\pi vk/d$. If the frequency is close to one of the resonant frequencies of the ion transition, then resonant excitation of a coherently scattered ion can occur in connection with small-angle reflection:

$$\omega_{mn}\approx 2\pi vk/d. \tag{5}$$

This phenomenon is analogous to resonant excitation of an ion channeled in the interior of a crystal.[10]

The second condition is formulated as follows. For coherent excitation of ions (by incident particles) it is necessary that the time during which the external field acts be less than the irreversible relaxation times:

$$l<vT_2<l_{\text{coh}}, \tag{6}$$

where l_{coh} is the distance over which coherent inelastic scattering of the ion occurs. The next condition for effective coherent collective excitation of ions assumes the form

$$\varepsilon_0\geqslant\hbar/d_{mn}T_2, \tag{7}$$

where ε_0 is the amplitude of the periodic electromagnetic field. When these conditions are satisfied a certain number of ions achieve a coherent collective state. Their number can be estimated as

$$N_1=N_0(1-\pi b_0^2Nd), \tag{8}$$

where N_0 is the number of beam particles and $b_0\geqslant(r_n^2+a^2)^{1/2}$, where r_n is the radius of the excited electron orbit.

5. GENERAL FORMALISMS

To describe coherent electromagnetic phenomena in an ion beam we can use the well-developed techniques of coherent optics in time-independent resonant media.[11] For this it suffices to consider the interaction process in the coordinate frame in which the longitudinal momentum vanishes. In this system we are working with a collection of ions which are almost at rest, coherently excited by the spatially periodic structure of the electromagnetic fields:

$$\mathbf{E}(t)=\hat{\varepsilon}(t)\cos\omega t, \tag{9}$$

where ω is one of the frequencies of the induced harmonics.

The resonant medium is initially in a state of thermodynamic equilibrium. Hence the statistical operator describing the state of the medium has the multiplicative property:

$$\rho(t_0)=\rho_n(t_0)\rho_T(t_0), \tag{10}$$

where $\rho_n(t_0)$ and $\rho_T(t_0)$ are the density operators describing the state of the ion and the heat reservoir respectively.

For an individual ion interacting with the external electromagnetic field under resonant coherent excitation conditions ($\rho=\rho_n$) we can write the equation

$$i\hbar\dot{\rho}=[H_0+V(t),\rho], \tag{11}$$

where $V(t)$ is the operator describing the interaction with the external field and the Hamiltonian describes the free particle. The formal solution of Eq. (11) can be written

$$\rho(t)=A\rho(t_0), \tag{12}$$

where $A=\exp(-iLt/\hbar)$ is the propagator and $L\rho=[H_0+V(t),\rho]$. Here we neglect the effect of the heat reservoir on the resonant ion because the interaction time is short.

After the field is turned off the resonant optical center interacts with the heat reservoir:

$$i\hbar\dot{\rho}=[H_0+U(t),\rho], \tag{13}$$

where $U(t)$ is the operator describing the interaction with the heat reservoir. The solution of (13) can be written in the form

$$\rho(t)=B\rho(t), \tag{14}$$

where $B=\exp(-iRt/\hbar)$ is the relaxation propagator and $R\rho=[H_0+U(t),\rho]$. In this treatment of coherent optical effects the density operator of the system at any time can be determined by the successive action of A and B. For example, after two identical pulses are applied

$$\rho(t)=BABA\,\rho(t_0). \tag{15}$$

The evolution of the density operator determines that of the induced particle angular momentum:

$$\langle \mathbf{d}(t)\rangle = S\rho(\mathbf{d}\rho). \tag{16}$$

The polarization vector can be evaluated by summing over all resonant ions, and in the case of a uniform medium we can write the macroscopic dipole moment (the polarization) as an integral:

$$\mathbf{P}(t)=n\int g(\Delta\omega)\langle \mathbf{d}(t)\rangle d\omega, \tag{17}$$

where n is the number of dipoles per unit volume (or area or length).

The next step is to solve the classical equation for the polarization of a medium when the right-hand side is known:

$$\Delta\mathbf{E}-\frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2}=\frac{4\pi}{c^2}\frac{\partial^2\mathbf{P}}{\partial t^2}+4\pi\ \text{grad div}\ \mathbf{P} \tag{18}$$

and the field and the intensity of the coherent collective radiation are calculated for a specific geometry.

After the sequence of operations (9)–(18) has been performed, these results can be transformed to the laboratory coordinate system by the corresponding transformations. The systematic procedure described above is a semiclassical approximation to the theory of coherent optical effects. It permits a relatively simple description of the kinetics of coherent transition processes and the phenomenon of coherent collective emission in various forms (free induction, photon echo, coherent spontaneous emission, etc.).

6. COHERENT OPTICAL EFFECTS

Using the formalism described above in Sec. 5 we look at the simplest model for the development of free induction and photon echos in an ion beam coherently excited by spatially periodic electromagnetic fields. By free induction and photon echo in optics we mean the process by which quantum systems radiate coherently from a coherent collective state. These effects result from the stimulated cooperative organization of elementary quantum emitters.

Assume that a set of monoenergetic two-level ions is extended along a single line perpendicular to the direction of motion, and that it interacts with a wiggler under conditions such that a quantum coherent state can result. In the beam it is quite easy to transfer every particle to the ground state, so we have $\rho_n(t_0)=1$. The solution of Eq. (13) for the density matrix of a two-level system with the interaction operator in the dipole approximation allows us to find the average value of the dipole moment induced by the interaction:

$$\langle \mathbf{d}(t)\rangle = \mathbf{d}_{21}\sin\theta(t)\sin\omega t, \tag{19}$$

where $\theta(t)=d_{21}/\hbar\int_0^{l/v}\varepsilon(t)dt$. If $\theta(t)\neq\pi$ holds we have coherent superposition; for $\theta(t)=\pi$ a state of complete inversion has been achieved.

The coherent collective state results from the self-consistent evolution of the whole set of particles and is described in the semiclassical approximation by the polarization vector. Taking into account reversible and irreversible relaxation processes for free decay we can write

$$\mathbf{P}(t)=N_1 d_{21}\sin\theta(t)\sin\omega t\exp(-t^2/4T_2^{*2})\times\exp(-t/T_2). \tag{20}$$

An oscillating macroscopic dipole radiates with an intensity which for $\theta=\pi/2$ is equal to

$$I(t)=N_1^2 I_0\exp(-t^2/4T_2^{*2})\exp(-t/T_2), \tag{21}$$

where I_0 is the intensity with which a single oscillator emits.

If we now consider the action of a new electromagnetic radiation pulse on the set of oscillating dipoles after a time $t_{21} > T_2^*$ with $t_{21}\ll T_2$, then by following the procedure of the general formalism we can derive a picture in which at time $t_{\text{echo}}=2t_{21}+l_1/v+l_2/v$ a macroscopic dipole forms again, radiating with power

$$I_{\text{echo}}(t)=N_1^2 I_0\sin^2\theta_1\sin^4\frac{\theta_2}{2}\exp\left(-\frac{t^2}{4T_2^{*2}}\right)\exp\left(-\frac{t}{T_2}\right). \tag{22}$$

This signal is called a photon echo; It is largest for $\theta_1=\pi/2$ and $\theta_2=\pi$, i.e., for $l_2=2l_1$.

In the laboratory coordinate system the photon echo is radiated at a distance $x_{\text{echo}}=2vt_{21}+l_1+l_2$. The free induction radiation and photon echo occur at the frequency $\omega=2\pi vn/d$ of the external field. For $d\sim$1–20 Å and $v\sim10^6$–10^7 m/s the radiation frequency includes the optical and x-ray wave bands.

For a relativistic ion beam coherent collective radiation has some distinctive properties. Thus, the intensity with which a set of relativistic particles radiate is greater by a factor of γ^4 (where γ is the Lorentz factor) than a nonrelativistic beam ($I\sim N^2 I_0\gamma^4$). The radiation is directed mainly forward within a small angle ($\theta<l/\gamma$) about the velocity vector and has a higher frequency ($\omega=2\omega_0\gamma^2$).

Let us consider one other variant of this scheme. Assume that the ion beam interacts with the laser beam moving in the opposite direction. In this case, in the rest frame of the ions the frequency of the light is increased by a factor of 2γ, and hence we can move into the region of higher coherent excitation frequencies $\omega_{21}=2\omega_0\gamma$, where ω_0 is the frequency of the laser radiation. By choosing laser pulses of the proper length in an appropriate sequence we can excite coherent collective states and observe coherent optical effects under new conditions. Estimates show that this technique allows deep quantum transitions to multicharged ions (in the x-ray band), and the output frequency is shifted into the γ-radiation wave band.

Note also that by combining a sequence of laser pulses with ion beams we can obtained a large variety of coherent interactions in the far-ultraviolet and x-ray regions of the spectrum. The problem of the coherent emission of

charged particles (including multicharged ions) in a resonant medium in the presence of a high-power optical pulse has been treated previously by one of us.[12]

7. CONCLUSION

The approach treated in the present work essentially allows familiar optical effects to be studied under new physical conditions. The allows the frequency of equivalent monochromatic radiation acting on a set of particles moving through a wiggler to be adjusted continuously by varying their speed. The concept opens up the possibility of dynamically regulating the properties of a resonant ion beam by means of the electromagnetic optical systems of the accelerator. The high intensity of the external radiation as well as the high sensitivity of existing photon detectors allows ion beams of quite low density to be used. And if all the conditions derived above are satisfied as accurately as possible, then we can expect to successfully observe coherent optical effects in an ion beam in the optical, x-ray, and γ-ray wave bands.

The practical implementation of this technique may have a number of interesting applications in atomic spectroscopy, beam microscopy, and wiggler microscopy. For example, the spectroscopy of deep transitions in multicharged ions is very problematical in existing optics. It is obvious that the concept presented here may be used to develop coherent spectroscopy of multicharged ions over a broad range of optical spectra.

Difficulties with efficient external pumping in the high-frequency region and difficulty in producing cavities for electromagnetic waves in the x-ray and γ-ray bands prevent conventional laser schemes from being used to develop x-ray and γ-ray lasers. Coherent collective emission from an ion beam can be used as a source of coherent adjustable electromagnetic radiation over a range of wavelengths including even the ultrashort-wavelength bands.

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