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Energetic electronic ensembles: new rules for the interaction of radiation with matter

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Abstract. The fundamental problem of the interaction of radiation with matter is considered for light intensities considerably larger than an atomic unit. By using intense femtosecond pulses in the UV spectral range, the coupling of radiation to matter is substantially modified. The usual weak coupling behavior characterized by the fine-structure constant $\alpha = 1/137$ has to be replaced by $Z^2\alpha$, where Z is the number of cooperatively acting electrons, which leads to anomalously strong coupling between radiation and matter.

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The development of technologies for the delivery and compression of power in materials is one of the oldest endeavors of mankind with an origin that predates the stone age. From the use of a wooden club to the contemporary production of vigorous thermonuclear environments, the achievable spatial power density (W/cm^3) has been advanced by approximately twenty orders of magnitude thanks to the availability of high-intensity light sources such as UV femtosecond laser systems [1] delivering intensities exceeding $10^{19}\,W/cm^2$.

An extremely general method of communicating power to matter involves the coupling of an externally generated field to the motion of electrons existing in the material. Common examples involve (i) the flow of a current of conduction-band electrons in a metal induced by an external electric potential and (ii) the excitation of valence-band electrons to continuum states in a semiconductor by the absorption of radiation having a quantum energy ($\hbar\omega$) exceeding the band gap energy (E_g).

Normally, the magnitude of the external field causing the excitation is very small compared with the naturally existing ambient field characteristic of the system, which typically has a value in the range of an atomic unit.

$$1\text{a.u.} \equiv \frac{e}{a_0^2} \approx 5 \times 10^9\,V/cm\,,$$

with e the charge of the electron and a_0 denoting Bohrs´s radius. In terms of an external light intensity such a field strength corresponds to $6 \times 10^{16}\,W/cm^2$ [2]. In addition the excited electrons respond as individual uncorrelated particles. Consequently, the behavior of the system is determined by a sum of interactions arising from weakly excited independent motions of individual electrons whose basic electromagnetic coupling strength is governed by the magnitude of the charge e of a single electron. This leads to a description of the strength of the interaction for these processes which is fundamentally based on the fine-structure constant α, a dimensionless physical parameter whose numerical magnitude is small compared to unity.

$$\alpha \equiv \frac{e^2}{\hbar c} \approx \frac{1}{137}$$

In the definition of α, c represents the speed of light and $\hbar$ denotes Planck´s constant, a basic parameter associated with the quantum theory of matter. It follows as a direct implication of the smallness of α that the ability of induced electronic motions to communicate power density to materials is subject to a limiting upper boundary whose magnitude is correspondingly small.

This basic limitation in the effective coupling strength, and the corresponding boundary on the power density, can be greatly surpassed if the independent uncorrelated electronic motions are replaced by induced motions of a sufficiently ordered form. This means that controlled power densities in materials can be advanced to a new, far higher and previously unreachable, level through the use of ensembles of electrons whose motions are well organized. The key factor is the cooperative additive action of the electrons comprising the ordered ensemble, an effect which vanishes in the case of independent uncorrelated interactions.

The use of the ordered electronic motions in appropriate materials can lead to an enormous and controllable enhancement in the effective strength of the interaction. Simple estimates [3–5] based on an analogy with atom–atom collisions [6] show that an ordered ensemble [7] comprised of Z electrons, which represents a system with a total charge of Ze, results in a modified coupling strength given by $Z^2\alpha$. Therefore, the effect of the ordered motion leads to the re-

placement of the customary coupling constant α by the larger value $Z^2\alpha$. Since there is preliminary evidence [3] that the number of participating electrons Z can fall in a range as high as 25–50, there is both the possibility that greatly increased effective coupling can be achieved ($Z^2 \sim 10^3$) and that a regime of anomalously strong coupling ($Z^2\alpha \gg 1$) can be dynamically produced [5] – a situation that is qualitatively different from the normal weak coupling condition ($\alpha \ll 1$). Therefore, an entirely new regime of strengthened physical interaction arises.

An electromagnetic coupling with this augmented characteristic strength ($Z^2\alpha$) could serve many highly important applications. Potential areas of use include new simple methods for the accurate micromachining of solid state surfaces, the production of practical high-brightness X-ray sources for advanced modalities of biological microimaging [8], and new unconventional forms of nuclear energy production. Apparently the use of the ordered electronic ensembles dramatically changes the rules governing the coupling of radiation to matter. This change is produced by a modification of the effective coupling strength governing the interaction from α to $Z^2\alpha$ for an ensemble consisting of Z electrons. Furthermore, since the use of appropriate materials would make $Z^2 \sim 10^3$ possible, a huge enhancement can be obtained. In order to realize this strengthened coupling in practice, two questions are paramount. Specifically, what are the physical conditions necessary to produce the sufficiently ordered motions of the electronic ensembles and is there any existing experimental evidence indicating that these conditions can actually be achieved? As discussed below, recent experimental data furnish the answers to both. The necessary physical conditions are experimentally established and practical methods for their production do exist.

We now discuss in simple terms the nature of the ordered motion undergone by the electronic ensembles. Electrons bound to normal unexcited atoms and molecules represent compact distributions of charge. Their localization occurs in two forms. The wavefunction – the mathematical function that gives the quantum-mechanical description of the electrons – describes an electronic distribution which is compact in both coordinate space and in momentum space, the latter being the product of the velocity and the mass of the electron. The spatial distribution of outer weakly bound electrons in all materials is confined to a size in the order of a typical atomic dimension, the basic magnitude of which is known as the Bohr radius, a_0. However, since there exists a minimum quantum of action $\hbar$ (Planck´s constant) which, although small, is not zero, confinement in space requires through Heisenberg´s uncertainty principle a finite distribution in momentum space whose corresponding scale is given by $\sim \hbar/a_0$. Therefore, viewed in a phase space of two dimensions, x (space) and p_x (momentum), the initial state of such outer electrons in atoms and molecules generally takes the form shown as zone A in Fig. 1. We will designate this original unexcited state of the electrons as "state A".

An excited state can be produced from state A by interaction with an electromagnetic field. In particular, a strong and rapid acceleration of Z electrons in state A induced by the field changes the distribution function in phase space (state) in two ways. It is displaced in both space (x) and in momentum (p_x), hence, state A goes to the new location in Fig. 1 denoted as "zone B". Note that state B is drawn

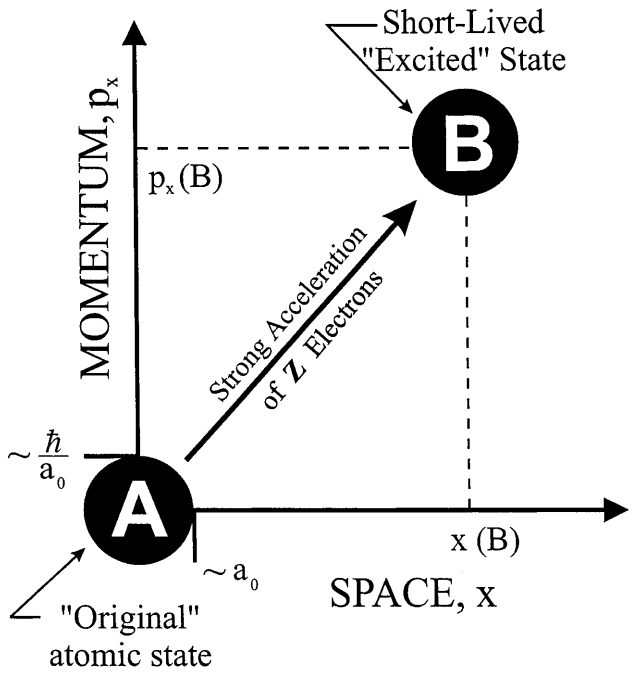

Fig. 1. Simple diagram of two-dimensional phase space (x, p_x) together with definition of zones A and B. Zone A characterizes the original, unexcited state of the electron, zone B a short-lived excited state. Further details in the text

with the compactness preserved, that is, the electrons still occupy only a small region of the available phase space ($x - p_x$ plane). However, this unusual condition certainly cannot exist for an indefinite time, since the electrons, now separated from the mutually attracting force of the positively charged atomic nucleus, which remains located at its original position corresponding to the center of state A, repel each other. Consequently, through this mutually repulsive interaction, the electrons will, in a sufficiently long time, disperse themselves throughout a much larger zone in the ($x - p_x$) phase space. Therefore, the compact distribution given by zone B (state B) represents a short-lived, highly excited multi-electron state of the system. We will designate the lifetime of this unusual many-electron well-ordered excited state as τ_B. This means that if the picture given in Fig. 1 is valid, the acceleration impressed on the Z electrons in state A must be strong enough and applied sufficiently rapidly to cause the transition from state A to state B to occur in a time comparable to or less than τ_B. Since acceleration and force are proportional as described by Newton´s laws, a statement that also holds in quantum theory, we see immediately from the morphology of Fig. 1 that the externally applied field (E_{ex}) must be large compared to the normal atomic ambient field (~ 1 a.u.) that binds the electrons to the atom. This conclusion follows directly from the form of Fig. 1, namely that $p_x(B) \gg \hbar/a_0$, and the proportionality of force and acceleration stated by Newton´s Laws. Therefore, we conclude that effective excitation of the electron ensembles requires that the applied external radiative field must be considerably larger than an atomic unit. It follows that the corresponding intensity of irradiation needed to generate the ordered motion of the ensembles is $\sim 10^{18}$ W/cm^2, a value which is a factor of 10^{19} higher than the intensity of the sunlight at the earth´s surface at noon (0.13 W/cm^2).

Estimates of τ_B and E_{ex} have been made in connection with experimental knowledge of phenomena that can be related to the conditions and processes described above. The physics of atom–atom and ion–atom collisions provides a basis for an estimate of E_{ex} which can be compared to the approximate range derived above. In particular, an analogy can be made with these collisional mechanisms [9] that gives the direct information on the competition between single-electron and multi-electron processes. Specifically, the observed properties of collisions between U^{32+} ions and Ne atoms indicate that multi-electron motions can dominate the interaction during the collisional encounter. This situation corresponds to the desired condition illustrated in Fig. 1 by the transition from state A to state B. The characteristics of this collisional process can in turn be interpreted in terms of the magnitude of the characteristic field needed to generate the multi-electron process. It is found from this analysis [9] that a value of E_{ex} for Xe atoms of ~ 10 a.u. – hence $> 10^{17}$ W/cm^2 – is a reasonable magnitude for the development of the appropriate many-electron motions in that system. This value is in good agreement with both the discussion of Fig. 1 above and experimental data recently obtained on X-ray production in Xe [9].

Two approaches have been used to provide estimates of the characteristic time τ_B based on available experimental data. They involve the Auger effect, a process in which an excited atom releases its energy by ejecting a fast electron, and the scattering of electron motions in a semiconductor (GaAs). Both of these processes occur through the direct naturally repulsive interaction of the electrons, the same mechanism that gives the decay of state B in Fig. 1. Information from the Auger process [7] puts τ_B in the 1–10 fs range (1 fs $\equiv 10^{-15}$ s). The experiments [10] concerning the semiconductor indicate a value of 1 fs $\leq \tau_B \leq$ 2 fs, a magnitude in agreement with the low end of the estimate derived from the Auger mechanism.

The establishment of the time τ_B in the range of a few fs has an important implication concerning the production of the ordered ensembles. Since a light wave presents a time-varying oscillating force on electrons, this suggests that radiation of a sufficiently long wavelength (λ_1) and, therefore, a sufficiently long period T_1 such that $T_1 > \tau_B$, cannot be effective in energizing the ensembles in a sufficiently compact form. Therefore, sufficiently long wavelength radiation will not exhibit the enhanced coupling and corresponding strong X-ray generation. Conversely, radiation with a sufficiently short wavelength (λ_2), so that the opposite condition $T_2 < \tau_B$ holds, can be effective in driving the ordered electronic ensembles and, consequently, will exhibit an enhanced coupling and correspondingly strengthened X-ray generation.

This dependence of the strength of the interaction on the wavelength of excitation provides a simple test for the existence of the envisaged enhanced cooperative many-electron coupling. Specifically, as the wavelength of the light is changed, it should be possible to pass through a transition zone which divides the two regimes of coupling; the short wavelength region should show strongly enhanced X-ray emission with respect to the long-wavelength zone. This is possible, since the X-ray emission serves as a good indicator of the strength of the coupling. Furthermore, the estimated values of τ_B given above suggest that the familiar ultraviolet (UV) and infrared (IR) regions of the spectrum may correspond to the two zones representing the two classes of limiting behavior.

Recently conducted experiments [11, 12] give confirming data directly supporting the picture presented above for the generation of an enhanced coupling arising from the production of energetic ensembles of electrons. The key experiments were conducted on Xe clusters with the use of powerful light sources in the UV (248 nm) and IR (800 nm) ranges. Clusters are small tightly packed groups of atoms, in this case estimated to be on average 20–30 atoms/cluster. It is useful to regard these systems as large molecules whose local atomic density resembles that of solid matter.

Xenon clusters are known to produce X-ray emission copiously in the keV range – corresponding to wavelengths as

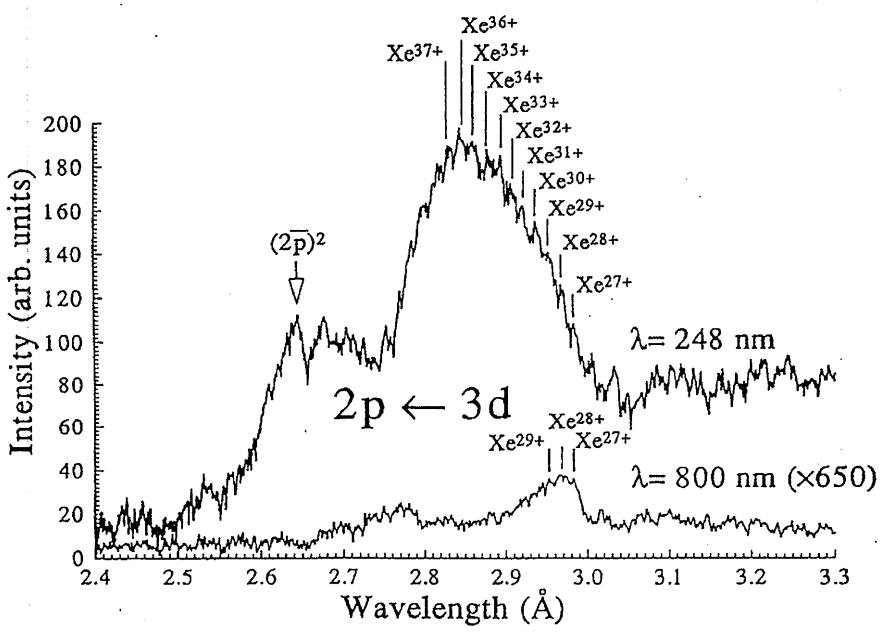

Fig. 2. Comparative Xe(L) spectra observed by irradiation of Xe clusters with wavelengths of 248 nm and 800-nm. The positions of the identified Xe^{q+} charge states are indicated. The ordinate of the 800 nm data is increased by a factor of 650 in order to enhance the visibility of the spectrum. The location of previously observed [3, 10, 12] double 2p vacancy species $(2\overline{p})^2$ is shown on the 248-nm data

short as ≈ 2–3 Å and ≈ 10–12 Å – under suitable irradiation by UV light [13]. This strong X-ray emission was viewed as evidence of the electronic ensembles [3, 5], but this interpretation lacked corroboration by corresponding data taken at other wavelengths. New data [11, 12] produced with 800-nm radiation provided the additional information needed, as described below.

In connection with the description of the ensembles given above, a good example of the relevant experimental findings is given by results concerning the generation of Xe(L) X-ray emission which occurs in the 2–3 Å region. The comparative Xe(L) spectra arising from irradiation at the two wavelengths used (248 nm and 800 nm) are illustrated in Fig. 2. The comparison of the two Xe(L) spectra reveals that both are very large in spectral width and there exists a considerable decrease in the peak intensity of the emission with the use of the infrared (800 nm) wavelength. In addition one observes very significant differences in the spectral characteristics of the two emissions. The spectrally integrated strengths (total X-ray emissions) differ by a factor of approximately 10^4. With regard to the comparison of the spectral structure, the 800-nm-induced signal exhibits only relatively low Xe^{q+} charge states ($q = 27$, 28, 29) and no double-vacancy $(2\overline{p})^2$ emission is detected. Double-vacancy $(2\overline{p})^2$ states are highly unusual excited states in which two inner-shell electrons are simultaneously absent. Previous studies [3, 5, 9] have indicated that the production of these exceptional double-hole species $(2\overline{p})^2$ is a characteristic signature of the presence of a mechanism of enhanced coupling. These two comparative findings, involving both the spectral strength and the specific structural differences in the spectra, are the expected characteristics of a greatly diminished amplitude for the multiphoton coupling with 800-nm radiation. Furthermore, both of these observations correspond exactly to the behavior predicted by the ensemble picture. Finally, within the framework of this model, the experimental findings [11, 12] at the two wavelengths used indicate an effective dephasing time for Xe clusters of ≈ 1–2 fs. Importantly, this range is consistent with the value expected from independent knowledge of electron–electron interactions. Specifically, the effective dephasing time found is in agreement with both the measured k-space scattering dynamics [10] of carriers in GaAs and the estimate based on the Auger process [7].

These experimental findings lead to the conclusion that the use of sufficiently intense femtosecond pulses of sufficiently short wavelength radiation enables one to modify substantially the coupling of radiation to matter, the strength of which is normally regarded as both constant and relatively weak. This new form of control arises from the use of energetic ensembles of Z electrons, systems which are compact in phase space and give an enhancement of the coupling by a factor of Z^2. Furthermore, since the factor Z can be varied by the choice of the material over the approximate range $1 \leq Z \leq 50$, an enormous and variable enhancement can be obtained. Consequently, these results point the way to a new more powerful and controllable method for the coupling (transmission and deposition) of radiation to all forms of matter. This new optically induced physical phenomenon is expected to lead to many new effects and applications based on the ability to increase controllably a basic physical parameter that was heretofore fixed at a small value.

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