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Optical Pumping between Levels of a Bistable State of Alkali Atoms Trapped in Rare-Gas Matrices

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The results of pseudopotential calculations of the interaction of an alkali atom with its rare-gas neighbors in a solid matrix are presented. They can explain an interesting effect which appears to be due to optical pumping between two equilibrium positions in a bistable trapping site.

In the course of laser-excitation experiments[1,2] to study the absorption and emission spectra of Na and K atoms trapped in rare-gas solids at 10°K, we have discovered an interesting effect which appears to be due to optical pumping between levels of a bistable configuration of the alkali-matrix system. On the basis of pseudopotential calculations of the alkali–rare-gas interaction, the effect can be explained in terms of a model in which the alkali atom moves from one stable position to another within a trapping site in response to optically induced changes in the alkali–rare-gas potential.

Matrix-isolated alkali atoms exhibit a number of distinct absorption bands associated with $^2S_{1/2} \rightarrow {}^2P_{1/2,3/2}$ transitions in atoms located in different trapping sites.[1-8] The absorption of white light by Na atoms trapped in solid Ar, as observed in our experiments (Ref. 1), is shown in Fig. 1. This scan is in agreement with earlier work[3] on Na and is similar to absorption profiles in other alkali–rare-gas systems.[2-6] The absorptions due to different trapping sites are labeled with capital letters. Also shown in Fig. 1 is the

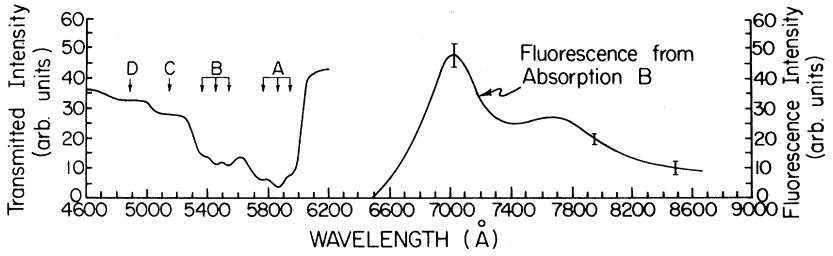

FIG. 1. A normalized scan of the absorption of white light by Na atoms trapped in solid Ar at 10°K. Also shown is the laser-excited fluorescence from absorption *B*.

fluorescence we observed when the sample was irradiated with a N_2-laser–pumped dye laser (pulse duration: 8 nsec; pulse energy: 100 μJ) tuned to a wavelength anywhere within the absorption triplet labeled *B*.

In optically thin samples the amplitude of the laser-excited fluorescence from absorption *B* decreased in time. No recovery of the signal amplitude in the dark was observed for time intervals up to 1 h after the laser was turned off. If the laser beam was focused onto a different spot on the sample, the signal amplitude immediately returned to its original value, and then again decreased in time. No similar bleaching effects were observed for absorption *A* or *C*. Absorption *D*, however, bleached very rapidly, the fluorescence amplitude dropping to zero after approximately twenty laser pulses. The fluorescence amplitude from absorption *D* also did not recover in the dark. The fluorescence profile was similar to that obtained from absorption *B*, but the rapid bleaching precluded detailed measurements. We immediately found that the bleaching of absorption *D* caused a recovery of the fluorescence amplitude from absorption *B* and vice versa.

We prepared Na-Ar samples in the dark and looked for laser-excited fluorescence from absorption *D*. There was none. Tuning the laser to absorption *B*, however, produced strong fluoresence signals. Tuning back to absorption *D*, we immediately found fluorescence. It is apparent that the site responsible for absorption *D* is not readily formed or is not occupied by Na atoms when the sample is grown, but is optically created or populated by irradiating absorption *B*. Once created by optical excitation, the state responsible for absorption *D* excited indefinitely in the absence of laser bleaching (we waited for times up to 1 h).

The same effect was observed in Kr and Xe matrices, and similar effects were also seen for matrix-isolated K atoms. As is the case for Na, the optically created absorption band for K atoms is the most blue-shifted of the absorptions, but optical pumping of the state responsible for this blue-shifted band occurs at wavelengths in a region overlapping both absorption *A* and absorption *B* for matrix-isolated K atoms. For both Na and K, the bleaching of absorption *D* is much more dramatic than for absorption *B*. It seems unlikely that the optically created state is a metastable excited electronic state of the matrix-perturbed alkali atom because of its extremely long life. Two other possibilities immediately come to mind: The change in the alkali–rare-gas potential when the alkali atom is excited to the *p* state either produces a shift of the configuration of the Ar atoms surrounding the alkali atom or causes the alkali atom to move from one stable position to another within a complex site. The latter seems the more likely explanation.

We have calculated[9] the energy-level structure of alkali atoms perturbed by rare-gas neighbors as a function of the alkali position in various hypothetical trapping sites, using the pseudopotential technique introduced by Baylis,[10] and extended by Pascale and Vandeplanque,[11] for the calculation of alkali–rare-gas dimer potentials. In these calculations, the Ar atoms remain fixed in position. Assuming that the core electrons in the alkali are unaffected by the rare-gas atoms, the Hamiltonian for the alkali valence electron is

$$H = H_A(\vec{r}) + \sum_n V(\vec{r}, \vec{R}_n) . \quad (1)$$

Here H_A is the Hamiltonian for the valence electron of the free alkali and V is a three-body potential between the valence electron, the alkali core, and one rare-gas atom. With the alkali ion at the origin, $\vec{r}$ is the electron coordinate and $\vec{R}$ is the position of a rare-gas atom. The sum is over the rare-gas atoms immediately surrounding the alkali in a given trapping site. In the Baylis model, $V(\vec{r}, \vec{R})$ is given by the sum of three terms:

$$V(\vec{r}, R) = F(\vec{r}, \vec{R}) + G(\vec{r}, \vec{R}) + W(\vec{r}, \vec{R}) , \quad (2)$$

where F is an attractive term due to electrostatic polarization of the rare-gas core by the valence electron and alkali ion, and G and W are pseudopotentials, derived from the Thomas-Fermi statistical model, which represent the repulsive energies between the valence electron and the rare-gas core and between the alkali and rare-gas cores. The alkali-matrix potentials are obtained by finding the eigenvalues of Eq. (1), using a limited basis consisting of eigenfunctions of H_A.

Our calculations show that the Na-Ar interaction is substantially repulsive for ground-state Na atoms at interatomic distances corresponding to a substitutional site. Indeed, a trapping volume corresponding to the removal of at least four Ar atoms is necessary for a Na atom to easily fit into a fcc Ar lattice. Calculations of the *s*- and *p*-state potentials for a Na atom trapped in such a site are in qualitative agreement (Ref. 9) with the observed emission and

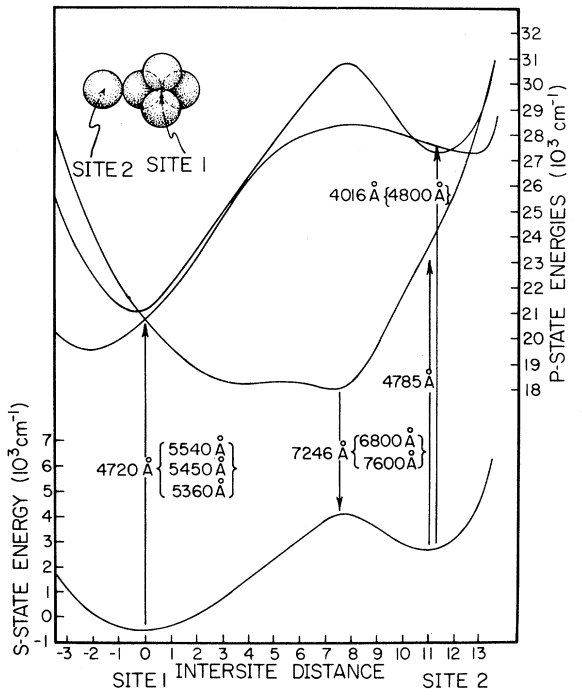

FIG. 2. A plot of the s- and p-state potentials for a Na atom in a bistable trapping site. The shape of the trapping volume is illustrated in the upper part of the figure. The spheres show the number and position of the Ar atoms that must be removed from the fcc lattice to create the trapping site. The potentials were calculated as a function of distance from one stable ground-state position to the other. The calculated transition wavelengths are noted; the corresponding experimental absorption and emission peaks are given in braces.

absorption spectra corresponding to absorption B. These calculations indicate that the smaller the trapping volume, the greater the blue shift of the absorption band. For these reasons we have taken as a model of a site that might exhibit bistable characteristics, a four-hole vacancy with an additional Ar atom removed adjacent to the large vacancy.

The s- and p-state potentials for an Na atom trapped in such a site are shown in Fig. 2, which also shows a sketch of the shape of the model-trapping volume. There are two stable positions for a ground-state Na atom; at the center of the four-hole vacancy (site 1) and at the center of the single-atom vacancy (site 2). Presumably site 2, which is repulsive, would not be readily occupied as the sample is grown from the vapor phase.

The p-state energy is split, corresponding to different orientations of the wave function's lobes, and these energy levels are strong functions of the atom's position, causing an atom excited to the p state to move off center. A computer search verified that the minimum p-state energy for this site corresponds to an alkali atom located at the entrance to the single-atom vacancy, one lobe inside the small hole, the other extending out into the larger volume.

Optical pumping would occur as follows. A Na atom at site 1 is optically excited to the p state at transition wavelengths corresponding to the p-s energy differences at this position. In the p state, it could move to the p-state minimum at the entrance to the single-atom vacancy. When it radiates back down to the s state, the emission wavelengths would be substantially red-shifted and it would be at a saddle point of the s-state potential. From this point it could either move to site 2 or back to site 1. Once in site 2, it could be optically pumped back to site 1 via the p state, either by the long-wavelength transition, which essentially coincides with the transition wavelengths for site 1, or by the shorter-wavelength transition to the higher p-state sublevels. The interaction with the lattice would presumably mix these levels and allow the atom to follow a path out to the entrance to the large hole.

This specific model for a bistable site should not be taken too seriously. The actual sites may not be straightforward vacancies in a regular fcc lattice, for the Ar matrix in the vicinity of an alkali atom may actually be amorphous. Furthermore, the pseudopotential calculations are not sufficiently accurate to quantitatively predict the observed emission and absorption wavelengths. Nevertheless, these calculations provide a ready explanation for an interesting optical-pumping effect and encourage one to believe that the optical spectra of matrix-isolated alkali atoms can be understood on the basis of the alkali–rare-gas interatomic potentials. It seems reasonable that more sophisticated calculations could be used to determine the structure of the trapping sites from experimental spectral data and that further experimental investigation of the bistable sites could be especially useful in this pursuit.

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Experimental Observation of Current Generation by Unidirectional Electron Plasma Waves

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A slow-wave structure was used to launch electron plasma waves traveling preferentially in one direction. The current generated by the waves was observed. The magnitude of the current can be estimated from momentum conservation in the wave-particle interaction process.

Plasma currents driven by traveling waves were first observed by Thonemann, Cowhig, and Davenport[1] in 1952, and the experiment was later repeated; see Bozunov *et al.*[2] and Demirkhanov *et al.*,[3] also Fukuda.[4] In principle, any wave with a net momentum can generate a current via any damping mechanism caused by the charged particles in the plasma. Ion cyclotron waves have been used to produce a current in the Princeton Model-C stellarator,[5] and current generation by whistler waves[6] has been reported recently. The application of rf-driven current to steady-state tokamak reactors has been considered by a number of authors.[7-10] If the current is carried by the main-body electrons, the rf power becomes too large to be practical.[7,10] Recently it was proposed to generate a current carried by the electrons in the tail of the distribution function[10] via Landau damping of lower-hybrid waves.[11,12] This idea relies upon the momentum transferred from the waves to the tail electrons and therefore requires unidirectional waves which can be generated by 90° phased metal plates or wave-guide arrays.[13] In this paper I report the first experimental observation of current generation by the unidirectional electron plasma waves via wave-particle interaction. Electron plasma waves are called lower-hybrid waves when the wave frequency is near the lower hybrid frequency. These waves are attractive to magnetic fusion research due to the desirable features of the launching structure (phased wave-guide arrays) and the readily available technology to produce the necessary rf power corresponding to the tokamak reactor parameters.

Consider the electron plasma wave which obeys the following dispersion relation:

$$\omega = \frac{k_z}{k} \frac{\omega_{pe}}{(1+\omega_{pe}^2/\omega_{ce}^2)^{1/2}}, \tag{1}$$

where k_z is the wave number along the magnetic field; k is the total wave number; ω, ω_{pe}, and ω_{ce} are the wave frequency, the electron plasma frequency, and the electron cyclotron frequency, respectively. Let E_0 be the electric field amplitude of the wave and let ϵ be the plasma dielectric function. The parallel component of the wave momentum density is

$$P_z = \frac{E_0^2}{16\pi} \frac{\partial}{\partial \omega} (\omega\epsilon) k_z = \frac{1}{8\pi} E_0^2 \frac{k_z}{\omega} \left(1 + \frac{\omega_{pe}^2}{\omega_{ce}^2}\right). \tag{2}$$

If the wave momentum is absorbed by the electrons, it will generate a current density

$$J_z = \frac{e}{m} \frac{E_0^2}{8\pi} \frac{k_z}{\omega} \left(1 + \frac{\omega_{pe}^2}{\omega_{ce}^2}\right), \tag{3}$$

where e/m is the electron-charge-to-mass ratio. In a toroidal plasma, the resonant electrons moving parallel to the magnetic field are confined. The wave-generated current will increase as long as there is a nonzero slope in the electron velocity distribution function for Landau damping until electron-ion collision transfers the electron momentum into the ions. In linear machines, the resonant electrons leave the plasma through the end of the machine before electron-ion collisions occur. Consider a pulsed wave packet of power P_w and pulse length τ. If the wave is completely damped by electrons, the total charge that leaves