for shallow acceptor to be considerably shorter. This is consistent with the interpretation given here that the stimulated echo arises predominantly from the indium sites with $g\mu_B H \ll \delta$.

The results of the present investigation can be summarized as follows. (i) The generation of a backward-wave phonon due to the parametric mixing of a forward wave and a microwave field has been detected in indium-doped silicon. (ii) The backward-wave phonon spectrum as a function of magnetic field has been measured and used to identify the source of the nonlinearity as resonance transitions within the neutral-acceptor ground state. (iii) The frequency, magnetic field magnitude and orientation, microwave power, and temperature dependences are all consistent with this interpretation. (iv) The narrow peak at $H = 0$ (Fig. 1) is not yet understood and there is some evidence that at higher temperatures ($T \simeq 10$ K) a relaxation interaction dominates over the resonance process. (v) The dependence of the strain and electric field matrix elements on the zero-field splitting reduces the inhomogeneous broadening, making possible the resolution of individual transitions. (vi) The spin-lattice relaxation time was measured at 1.35 K using a new form of transient-population-grating spectroscopy.

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Marginal Fluctuations in a Bose Glass

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The possibility of Bose condensation into the lowest-energy, extended, Hartree-Fock eigenstate in the presence of a random potential is studied. Using the existing folklore about Anderson localization, and adding interactions, we find that the fluctuations are logarithmically divergent and that stable broken symmetry is impossible for $n \geq 2$.

In recent years there has been considerable interest in the understanding of collective phenomena in random systems such as spin-glasses and disordered magnets. One approach[1] is to do perturbation theory simultaneously in both the disorder and the interactions between modes which are independent in mean-field theory. These calculations are only valid near four dimensions in systems with sufficiently small interaction strength and disorder.[2] They cannot shed light directly on the possibility of other sorts of critical points or more exotic behavior outside this domain. A particular example of this sort of behavior is a spin-glass transition to a state characterized by an Edwards-Anderson order parameter. This possibility has been studied by several authors who have

examined various models in mean-field theory.[3-5] Despite several years of intense theoretical effort, the stability and fluctuations of the Edwards-Anderson order are not completely understood in any of the models that have been studied.

In an effort to circumvent this impasse, we investigate an alternative theoretical approach that is appropriate for systems with moderate or strong disorder. It is based on a picture introduced by one of us[6] several years ago, in order to describe certain features of spin-glass materials. This was before any strong experimental evidence for a sharp transition[7] existed. The basic idea was to identify the normal modes of a random spin system with the localized eigenstates of single-particle localization theory.[8] The apparent failure of the thermodynamic functions to exhibit any sharp behavior was explained by the absence of phase transitions in finite systems. The current use of this picture[9] would then associate the spin-glass transition with the mobility edge while the random, frozen magnetization pattern corresponds to the first extended eigenstate of the exchange matrix.

In this Letter we describe explicit model calculations for the suppression of instabilities corresponding to localized modes, the possible phase transition at the mobility edge, and the destruction of long-range order in the first extended state by generalized Goldstone modes. The model may be applied to random spin systems, dirty ferroelectrics,[10] or a Bose fluid in a random potential. In the first two cases one would look for a phase transition as the temperature is lowered. In the latter case we envision raising the chemical potential. We will use, for simplicity, the language of the spin-glass transition.

The model is described by an effective Hamiltonian with a random exchange term and a quartic, on-site interaction. We write[11]

$$Z=\int \delta^3 S \exp\{-T^{-1}[\tfrac{1}{2}\sum_{x_1x_2}(T\delta_{x_1x_2}-J_{x_1x_2})\vec{S}_{x_1}\cdot\vec{S}_{x_2}+\tfrac{1}{4}U\sum_{x_1}(\vec{S}_{x_1}^{\,2})^2]\}, \tag{1}$$

where $\vec{S}_x$ is a soft n-component spin on the site x. For Bose systems the matrix $T\underline{1}-\underline{J}$ is replaced by $\underline{H}-\mu\underline{1}$, where $\underline{H}$ is a single-particle Hamiltonian, and μ is the chemical potential. Note that when $\underline{J}$ is translationally invariant, (1) exhibits a mean-field transition when T equals the largest eigenvalue of $\underline{J}$.

We transform to the basis, $|m\rangle$, of eigenstates of $\underline{J}$ so that

$$Z=\int \delta^3 S \exp\{-T^{-1}[\tfrac{1}{2}\textstyle\sum_m (T-J_m)\vec{S}_m^{\,2}+\frac{1}{4}\sum_{m_1m_2m_3m_4} U_{m_1m_2m_3m_4}(\vec{S}_{m_1}\cdot\vec{S}_{m_2})(\vec{S}_{m_3}\cdot\vec{S}_{m_4})]\}, \tag{2}$$

where

$$U_{m_1m_2m_3m_4}=u\textstyle\sum_x \psi_{m_1}(x)\psi_{m_2}(x)\psi_{m_3}(x)\psi_{m_4}(x) \tag{3}$$

and the $\psi_m(x)$ are the eigenfunctions of $\underline{J}$. Although the explicit diagonalization of the matrix $\underline{J}$ is generally impossible, the general features of its spectrum are fairly well understood.[12] We will assume that the spectrum has a mobility edge which divides extended states from localized states. The specific properties of these states will be discussed as they are needed.

Stability requires that the eigenvalues of the susceptibility matrix $\chi_{m_1m_2}^{\mu\mu}=T^{-1}\langle S_{m_1}^{\mu}S_{m_2}^{\mu}\rangle$ be nonnegative. The Gaussian approximation to (2) satisfies this requirement as long as the temperature is greater than the largest eigenvalue of $\underline{J}$. In a nonrandom ferromagnet this eigenvalue corresponds to a uniform mode. When the temperature becomes lower than this value, stability is restored by means of a finite magnetization in this eigenstate. (In the Bose fluid, this is the macroscopic occupation of the $k=0$ state.) As noted in Ref. 6, this mechanism cannot apply to the state with the largest eigenvalue of J since it is localized. This is perhaps even more obvious for the Bose system. A macroscopic number of particles in a localized volume will result in a prohibitive cost in repulsive energy.

These ideas find an explicit realization in a Hartree-Fock approximation to our model. We define the Green's function, $G=T\chi$. The Hartree-Fock Hamiltonian is

$$\langle x\mu|H^{\mathrm{HF}}|x'\mu'\rangle = [J_{xx'}-u\textstyle\sum_\lambda G_{xx}^{\lambda\lambda}\delta_{xx'}]\delta_{\mu\mu'}-uG_{xx}^{\mu\mu'}\delta_{xx'}, \tag{4}$$

where $G=(1-H^{\mathrm{HF}}/T)^{-1}$. Rotational invariance ensures that the self-consistent eigenstates, $|\tilde{m}\alpha\rangle$, are n-fold degenerate. We note that the corresponding wave functions, $\Psi_{\tilde{m}}^{\alpha\mu}(x)$ need not have trivial structure in the component indices; that is, the polarization may vary with position. We can always represent $\Psi_{\tilde{m}}^{\alpha\mu}(x)$ as the product of a rotation matrix $R_{\tilde{m}}^{\alpha\mu}(x)$ with a scalar function $\Psi_{\tilde{m}}(x)$. In this basis,[13] the Hartree-Fock Green's

function is

$$G_{mm}=\{1-J_m/T+(n+2)/T^{-1}\textstyle\sum_n U_{mmnn}G_{nn}\}^{-1}, \quad (5)$$

where we have dropped the tilde and the spin indices.

For highly localized states all terms other than the self-interaction term may be neglected, and G_{mm} can be obtained explicitly:

$$G=\frac{J_m-T+[(J_m-T)^2+4(n+2)U_{mmmm}T]^{1/2}}{2(n+2)U_{mmmm}}, \quad (6)$$

which remains positive. As we near the mobility edge, terms other than those with m equal to n make more sizable contributions. But as long as the state $|m\rangle$ is localized its contribution is always present and finite. Therefore, our stability argument is still valid since the presence of G in the denominator of (5) limits the growth of the expression as T is lowered, and stability is maintained.

Let us briefly return to the effective Hartree-Fock eigenstates. We can say that the spectrum is distorted from its noninteracting form so that all of the eigenvalues remain less than T. Of course, the basis that we are using is also temperature dependent, but it should have the same general features as the original. Let us examine the situation near the mobility edge somewhat more closely. For localized states with long localization lengths, we expect the general structure of the self-energy to be similar to that for extended states of a finite system whose size is roughly the localization volume λ^d. That is, the difference between the self-energy for an extended state near ϵ_c and that for a localized state just the other side of the edge is the difference between an integral and a Riemann-sum approximation to that integral. The typical discrepancy can be measured by the value of a single term, such as the one retained explicitly in (6). From this we conclude that the extra self-energy shift given to localized states near ϵ_c is proportional to $u\lambda^{-d}\propto u(\epsilon-\epsilon_c)^{\nu d}$ where ν is the localization length exponent. Thus, for $\nu>d^{-1}$ localized states right near the edge do *not* get pushed across it, although states farther away may be. Singular behavior in ρ^{HF} is averted since the states will become extended if the density becomes too great. Therefore, since calculations show that $\nu>1/d$,[14] we expect the mobility edge to retain its identity with a continuous density of states as one crosses it. The distortion of the spectrum is indicated in Fig. 1(a) and 1(b).

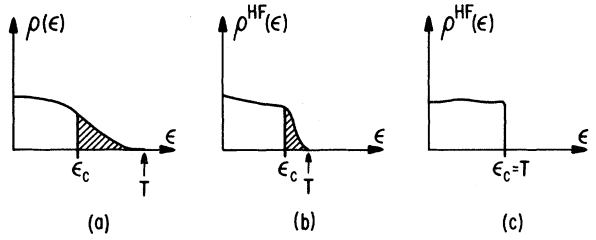

FIG. 1. Hartree-Fock density of states at three different temperatures (schematic): (a) For high T, $\rho^{\mathrm{HF}}\approx\rho$ = density of eigenvalues of $\underline{J}$; (b) for intermediate T, tail of localized states moves to keep to the left of T; (c) for T reaching the mobility edge, no localized states remain.

If and when T ever reaches the mobility edge [Fig. 1(c)], the situation is drastically changed. At this point all of the Hartree-Fock states are extended, and every term in the sum in denominator of (5) will make a contribution of order N^{-1}, where N is the number of spins in the system. The argument used above no longer guarantees stability.

Can we then expect a phase transition below which the first extended eigenmode acquires a spontaneous, frozen magnetization? If $n=1$, we see no reason why this should not be the case, in agreement with Anderson and Pond.[9] As T is lowered below the critical temperature, the macroscopic occupation of the state at the mobility edge stabilizes the fluctuations of all the other states, just as in the Ising model. If, however, $n>1$, we must reckon with Goldstone modes. Just as in the nonrandom problem, the equation of state, which determines the magnitude of the order parameter, implies the divergence of the transverse fluctuations of the order-parameter mode [Fig. 2(a) and 2(b)]. Thus $G_{mm}\propto(\epsilon_c-J_m^{\mathrm{HF}})^{-1}$, where J_m^{HF} is the Hartree-Fock eigenvalue of mode $|m\rangle$ and ϵ_c is the mobility edge. When we compute the self-energy due to a loop of these Goldstone modes,

$$\Sigma_m{}^{\perp}\propto\textstyle\sum_n U_{mmnn}/(\epsilon_c-J_m{}^{\mathrm{HF}})\propto\int_{-\infty}^{\epsilon_c}\frac{\rho^{\mathrm{HF}}(\epsilon)}{\epsilon_c-\epsilon}d\epsilon, \quad (7)$$

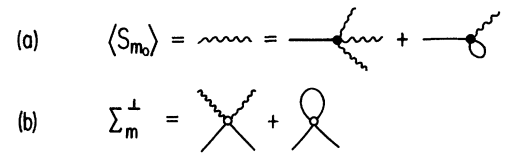

FIG. 2. (a) Equation for order parameter $\langle S_{m_0}\rangle$. ($|m_0\rangle$ is the first extended HF eigenstate.) (b) Equation for transverse self-energy in presence of finite $\langle S_{m_0}\rangle$.

we obtain a logarithmic divergence, because the Hartree-Fock density of states $\rho_1^{HF}(\epsilon)$ is slowly varying near ϵ_c. Thus, the collective modes destroy the order; the first extended eigenmode cannot be macroscopically occupied. The instability is qualitatively like that which occurs in two-dimensional nonrandom systems, where $\rho(\epsilon)$ is also constant near the band edge. Our picture can thus be viewed as a kind of "dimensional transmutation" at a critical temperature where the critical fluctuations change from something essentially zero dimensional in character to something rather two dimensional. We note that this result does not depend on dimensionality except insofar as it involves the existence of the mobility edge. We note that some recent work[15] indicates that there is no true mobility edge for $d = 2$.

This picture has several attractive features in application to spin glasses. First, the near divergence of the susceptibility associated with the localized states finds a natural physical interpretation in terms of spin clusters.[6,16] As far as we know, cluster effects are universal in spin glasses. Furthermore, the way the localized states get pushed across the mobility edge and become extended by mixing with the already extended states provides some theoretical underpinning for the picture of intercluster coupling introduced in the phenomenological cluster theory.[17] Secondly, one would like to associate the fact that the potential order is marginally unstable with the observed fragile nature of the spin-glass state.[18] Third, the constant density of states near the mobility edge accounts for a linear specific heat. Nonlinear behavior of the specific heat at very low temperatures finds an interpretation in terms of a picture where T has not quite reached ϵ_c. And finally, this approach allows us to study a kind of random ordering while avoiding the pathologies of the Edwards-Anderson order.

We have to admit that we do not know the domain of validity of this theory. While our approach formally treating the randomness exactly and doing perturbation theory in the anharmonicity of the fluctuations) appears to give a universal result, independent of the type or degree of randomness, it is obvious that one can instead do a better (nonperturbative) job on the anharmonicity and attempt to treat the disorder perturbatively. References 1 and 2 are just this sort of calculation, and they give a qualitatively different sort of result. Empirically, it is apparent that disordered magnets comprise several universality classes, depending on n, d, and the type and degree of randomness. Here we only hypothesize the existence of a class (including spin-glasses) to which the foregoing description applies. Ordinary, slightly disordered ferromagnets presumably belong to a different class, described by Ref. 1 and 2. Unfortunately, to find out into which class a given system falls would require a reliable treatment of both disorder and anharmonicity, and no method of doing such a calculation is known.

Of course, our argument only rules out broken symmetry characterized by $\langle S_{m_0} \rangle \neq 0$. It cannot tell us (just as its counterpart for nonrandom systems cannot single out the special case $n = 2$ for $d = 2$) whether there is a transition to some state without this kind of order. The answer to this question depends on understanding topological excitations in random systems, which may be quite different from their nonrandom counterparts.[19]

It would be attractive to apply this picture to nonuniform He^4 films,[20] but the situation is clouded by the possible nonexistence of a mobility edge for $d = 2$. Nevertheless, one does expect[15] a rather narrow crossover from exponential to logarithmic behavior in this case, so one might still be able to observe effects of virtual order and its fluctuations which were qualitatively similar to what one would find if the edge were sharp.

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Surface Coherent Anti-Stokes Raman Spectroscopy

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We demonstrate here that nonlinear mixing of four surface-plasmon waves can be used to probe the Raman resonances of liquids. The results are in good agreement with theoretical prediction. The technique should be useful for surface studies.

The propagation of surface electromagnetic waves on solids and their applications have recently attracted considerable attention.[1] They have been used to study adsorbed molecules[2] and overlayers on surfaces,[3] to probe phase transitions,[4] etc. In most cases, linear optics is employed in the excitation and detection of the surface waves. Observations of nonlinear optical processes involving surface electromagnetic (EM) waves have been rather rare. Simon, Benner, and Rako[5] have used the linearly excited surface-plasmon wave on metal films to generate a bulk second-harmonic wave. DeMartini *et al.*[6] have used the mixing of two bulk waves to generate a surface EM wave, and have used the mixing of a bulk wave and a surface wave as a means to detect the surface wave. Since high-intensity surface EM waves can be readily excited, one would expect that pure surface nonlinear optical effects (i.e., all input and output optical waves are surface waves) should also be easily observable. In this paper, we present the first results of such an experiment on the mixing of four surface-plasmon waves.[7]

The process we have been studying is the surface coherent anti-Stokes Raman spectroscopy (CARS). Two surface-plasmon waves at ω_1 and ω_2 propagate on the plane boundary surface between a metal and a dielectric medium with wave vectors, $(\vec{k}_1)_{\parallel}$ and $(\vec{k}_2)_{\parallel}$, respectively, parallel to the surface. These waves interact on the surface via the third-order nonlinearity in the medium to produce a third-order nonlinear polarization at $\omega_a = 2\omega_1 - \omega_2$ which in turn generates a surface anti-Stokes plasmon wave at ω_a. This anti-Stokes generation will be phase matched if $(\vec{k}_a)_{\parallel} = 2(\vec{k}_1)_{\parallel} - (\vec{k}_2)_{\parallel}$, and will be resonantly enhanced if $\omega_1 - \omega_2$ approaches the resonant frequency of some excitation in the medium. Therefore, just like bulk CARS, the surface CARS can also be used as a spectroscopic technique to study the resonances in a medium.

The theory of surface CARS is a straightforward extension of the theory on nonlinear generation and detection of surface polaritons developed earlier.[8] Suppose the Kretschmann geometry[9] [Fig. 1(a)] is used for excitation of the surface plasmons. The dispersion relation of the surface