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¹²Our molecular dynamics values of D , Fig. 1, are accurately represented by Eq. (2) with $D = 0.86$ and $Q = 2.1$. The mean free path was defined as the total distance traveled by a particle along one Cartesian coordinate divided by the number of reversals in the corresponding velocity component, and our values are approximated by the polynomial $\Lambda = 0.0829 - 0.258T + 0.18588T^2$. The difference in chemical potential between the supercooled liquid and the crystal is $\Delta\mu = T \int_T^{T^m} (E_L - E_c) dT/T^2$, where

$$E_L = -7.1847 + 4.1896T + 0.2702T^2 + 0.78396T^3$$

and

$$E_c = -7.4487 + 3.0127T + 0.24391T^2 + 0.55607T^3 + 1.7928T^4 - 6.2339T^5$$

are polynomials representing the internal energies (per atom) of the liquid and crystalline phases, respectively. The expression relating $\Delta\mu$ to the internal energies is derived from the Gibbs-Helmholtz relation, and is valid only when the pressure is zero.

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Monochromatic Phonon Generation by the Josephson Effect

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ac-Josephson junctions of tin and lead were found to emit monochromatic phonons with Josephson frequency, rather than thermal phonons. This is a new tunable phonon source with high efficiency and prospectively extremely high frequency resolution.

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We have investigated the phonon emission spectra of ac-Josephson junctions¹ with a stress-tuned phonon spectrometer² whose resolution was greatly improved.³ Surprisingly, we did not find the expected broad spectral distribution of phonons, but rather a sharp monochromatic peak. This peak is coincident with the Josephson frequency $\hbar\omega = 2$ eV and, accordingly, is tunable by the voltage. Thus, we have discovered a new tunable source of truly monochromatic phonons which is, at least with respect to frequency resolution, superior to the previous method of phonon "bremsstrahlung."⁴ Since the phonons are excited by the electromagnetic waves in the junction, the new effect must also have some bearing on the noise resistance of Josephson junctions.

The Josephson junction was evaporated onto a silicon crystal of dimensions $2 \times 4 \times 15$ mm³, doped with 5×10^{13} cm⁻³ boron acceptors [see inset (a) of Fig. 1]. The phonons emitted by the junction traveled ballistically through the crystal and were detected by a broadband detector (aluminum junction) on the far side. The boron acceptors have a fourfold-degenerate ground state

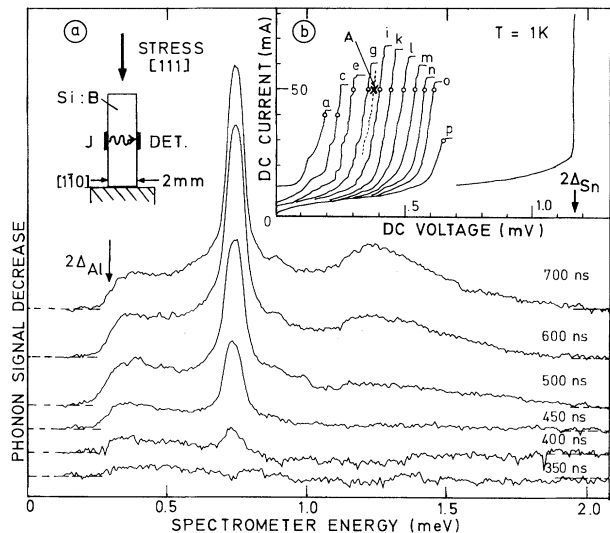


FIG. 1. Time evolution of the phonon spectrum for a fixed Josephson frequency. Insets: (a) geometry of spectrometer crystal, J is the Josephson junction; (b) I - V characteristic of the Sn junction at various magnetic fields.

which was split into two Kramers doublets by uniaxial stress perpendicular to the phonon path. The splitting gave rise to weak scattering of the phonons with the resonance frequency, thereby diminishing the total detector signal. The decrease of the detector signal as a function of the variable stress splitting is therefore a replica of the emitted phonon spectrum. The stress scale was calibrated against the energy scale by using the phonon bremsstrahlung.^{3,4}

To enhance the generation of electromagnetic waves, we used the resonance "bump" discovered by Eck, Scalapino, and Taylor.⁵ A weak magnetic field (< 10 G) was applied parallel to the junction plane to impose a spatial phase variation on the order parameter with a certain wavelength λ . When the Josephson frequency $f = (2 \text{ eV})/h$ matches the condition $f\lambda = \bar{c}$, where \bar{c} is the speed of electromagnetic waves in the junction, resonant excitation occurs which manifests itself by a steep current rise in the I - V characteristic. This is shown for a tin junction in Fig. 1, inset (b), for various magnetic fields.

The phonon nature of the emitted spectrum at, say, operating point A [Fig. 1, inset (b)] is best proven by the time of flight. If the usual pulse technique was used, however, there was inevitably some ringing on the pulse top which modulated the Josephson frequency and hence blurred the spectra. Therefore, we used a dc voltage to determine the operating point. Time resolution was nevertheless achieved by switching the dc voltage off within 30 ns, and observing the detector signal *thereafter*. The detector signal lasted for some 400 ns, thus reflecting the phonon time of flight, before it dropped. The amount by which the signal had dropped at a given time after switching is due to all phonons with times of flight shorter than this. The drop was measured at various delay times by a dual-gate boxcar average as a function of stress. The resulting spectra are shown in Fig. 1.

The spectrum starts rising after a delay time of 400 ns in fair agreement with the time of flight of fast transverse phonons which are dominant in the $[1\bar{1}0]$ direction as a result of phonon focusing. After 450 ns, the salient feature is a sharp peak at 0.74 meV which is exactly twice the bias voltage. The peak width equals the spectrometer resolution,³ indicating an extremely narrow bandwidth of the generated phonons. In addition to the peak, there is a broad background of phonons which is cut off below the threshold of the aluminum detector, $2\Delta_{Al}$. After 500 ns, a broad sec-

ond peak develops at 1.2 meV. This is clearly due to recombination phonons which are delayed by reabsorption.⁶ No indication of a direct electromagnetic signal was found, nor was it expected, because of the strong impedance mismatch of an extended junction to free space.

If the bias voltage was shifted by the magnetic field, the monochromatic phonon peak was tuned accordingly. This is shown in Fig. 2 for a delay time of 700 ns. The height of the peak is apparently increasing with the voltage (see traces *a-i*), mainly as a result of the spectrometer weight function.³ A dramatic drop of the peak height occurs, however, in going from trace *i* to *j*. Simultaneously, the 2Δ -phonon peak is increased. This signals the onset of photon-assisted single-particle tunneling⁷ at $eV = 2\Delta/3$ as a loss channel for the photons which is competing with the monochromatic phonon generation. The photon-assisted tunneling process creates quasi-particles which in turn recombine under 2Δ -phonon emission.⁷ At higher voltages, the Josephson peak itself eventually merges into the 2Δ phonons.

At low bias voltage, traces *a-e* in Fig. 2, there is also a peak at the second harmonic of the Josephson frequency. This undergoes reabsorption on the following traces. The second-harmon-

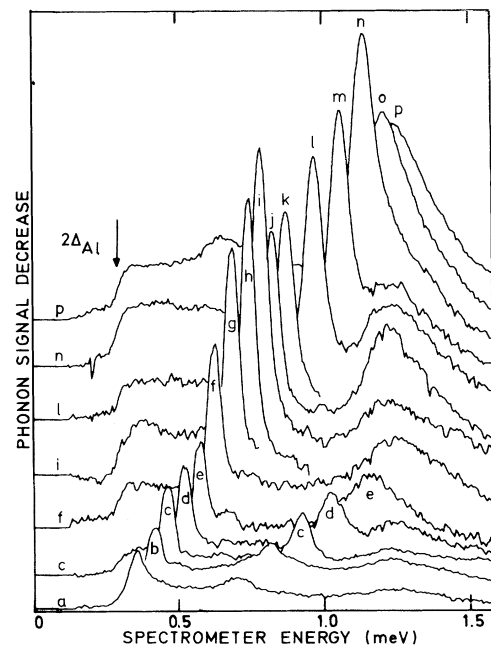


FIG. 2. Spectra for various bias voltages, correspondingly displaced in the vertical direction. The operating points are indicated in Fig. 1, inset (b).

ic generation is much weaker for junctions with higher tunneling resistance. No further harmonics have been found yet.

If the dc current was increased while the bias voltage was held fixed by readjusting the magnetic field, the peak height of the Josephson phonons increased accordingly; see Fig. 3. The dc current scales with the electromagnetic wave amplitude.⁵ At the same time, the constant dc voltage implies a constant amplitude of the ac-Josephson current.⁸ Hence, the phonons must arise from 1:1 conversion of the electromagnetic waves, rather than from direct coupling to the ac-Josephson current.⁹

The background intensity of phonons also increased in Fig. 3 with the electromagnetic wave amplitude. This suggests that the greater part of the background is due to photon-assisted tunneling, harmonic generation with reabsorption,⁸ and subsequent quasiparticle relaxation and recombination, while a smaller part must arise from thermal quasiparticle tunneling. For practical applications, the background should be as small as possible. This requires the 1:1 conversion to be much stronger than the competing processes. This is the case for lead junctions; see Fig. 4.

To estimate the coupling strength of the 1:1 conversion process in Sn junctions, we use the fact that it is comparable, according to trace *j* of Fig. 2, to the photon-assisted tunneling process at the onset $V_{dc} = 2\Delta/3e$. The latter is quantitatively known¹⁰ and yields, in lowest order, the phonon output power

$$P_{\text{assist}} = 2\Delta (V_{ac}/4V_{dc})^2 I(2\Delta)/e,$$

where V_{ac} is the spatial average (rms) of the ac voltage, and $I(2\Delta)$ the tunneling current above the

gap voltage. The coupling strength can be conveniently expressed by the effective admittance $Y = P/V_{ac}^2$. With $I(2\Delta) = 0.5$ A, we have $Y_{\text{assist}} = 200 \Omega^{-1}$. Any 1:1 conversion mechanism must yield an admittance of the same order.

The most common way of conversion is the piezoelectric effect. Assuming that the oxide barrier is piezoelectric as a result of the growth process (preferential orientation, polar impurities, nonstoichiometry, etc.), one would obtain the acoustic power¹¹

$$P = A (\omega \bar{e} V_{ac})^2 / 4\rho v^3.$$

Here, A is the area of the junction, \bar{e} the piezoelectric constant, ρ the density, and v the sound velocity. The oxide thickness was assumed to be much smaller than the wavelength. With $A = 0.7$ mm², $\hbar\omega = 2\Delta/3$, $\rho = 7.3$ g/cm³, $v = 1.7$ km/s for tin, and taking $\bar{e} = 0.2$ A s m⁻² like in quartz, we obtain $Y \approx 250 \Omega^{-1}$, obviously the right order of magnitude.

For this mechanism one may expect the generation of coherent plane waves propagating perpendicular to the barrier, because the wavelength of the photons is much larger than that of the phonons. However, the oxide layer is far from being planar on the scale of the phonon wavelength, since the metal films look tarnished to the naked eye. In fact, we have preliminary results showing that the angular distribution rather corresponds to diffusive emission.

Compared with piezoelectricity, the conversion of the electrostatic transducer made up by the two films of the junction is negligible. The same holds for the conversion by the magnetic pressure of the electromagnetic waves, and for the Abeles mechanism¹² which relies on the momen-

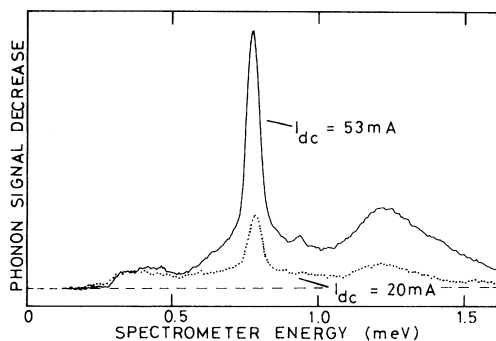


FIG. 3. Spectra for two different dc currents at $V_{dc} = 0.39$ mV. The monochromatic peak scales with the dc current, and hence with the electromagnetic wave amplitude.

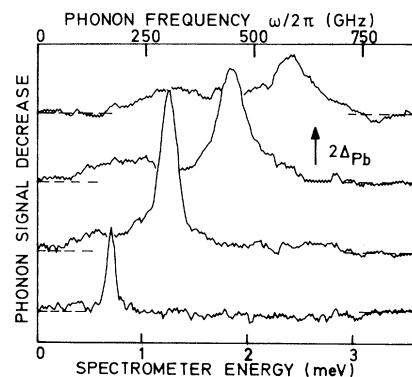


FIG. 4. Lead junction at various bias voltages. The background is smaller, and the frequency range is larger than for Sn junctions.

tum transfer of accelerated quasiparticles to the lattice.

There is a further possible excitation mechanism which is truly incoherent. If the oxide is considered to be an amorphous material, it should contain two-level systems which absorb the electromagnetic waves resonantly.¹³ The excited two-level systems will relax quantitatively via emission of phonons of the same frequency, because of the large phonon density of states. In this case, the absorbed photon power is¹³

$$P = 2\pi A \omega \mu'^2 n(E) V_{ac}^2 / d,$$

where μ' is the dipole moment, $n(E)$ the density of states of the two-level systems, and $d \approx 1$ nm the oxide thickness. In a dirty glass (BK7), $\rho^2 n(E) / 4\pi \epsilon_0 = 3 \times 10^{-4}$ was observed.¹³ Since the oxide was grown under fairly dirty conditions, we may use this value, and obtain $Y \approx 100 \Omega^{-1}$, again the right order of magnitude.

In conclusion, we have discovered a new source of tunable monochromatic phonons with frequencies up to the energy gaps of superconductors. The frequency resolution of the source should prove to be extremely high, because it depends only on the stability of the Josephson frequency. As yet, there are two possible ways to explain the effect quantitatively, namely, by the assumption of piezoelectricity or of two-level systems

in the oxide. A decision may be made by experiments on the angular distribution of phonons emitted by smooth junctions.

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Dynamics of Methane Monolayers Adsorbed on Graphite

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Pulsed-NMR measurements of methane on graphite have been performed between 45 and 105 K for 0.2 to 1.1 monolayers. Spin-relaxation times show first-order melting at 57 K, sensitivity to the hypercritical fluid, and continuous transitions where an incommensurate solid displaces the registered phase at both higher temperature and coverage. These measurements, plus activation-energy comparisons, provide a significant clarification of methane's high-temperature behavior.

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The widespread interest in adsorbed films on graphite is sustained by a continual unfolding of novel predictions and observations. In particular, registry and associated commensurate-incommensurate transitions (CIT's) have inspired studies of Potts model analogies, orientational phases, dislocation models, domain-wall configura-

tions, possible low-temperature liquids, and chaotic phases for coverage-driven CIT's.¹ In addition, transitions from a registered phase into a higher-temperature incommensurate (floating) solid have been proposed.² The phase diagram of adsorbed krypton³ possesses a well-developed registered phase which melts directly into a two-